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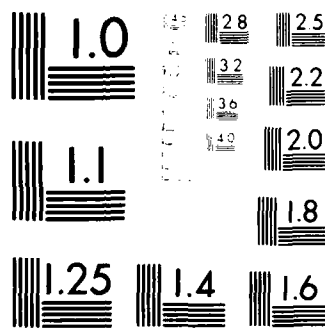
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FUELS COMBUSTION RESEARCH

For the Period 1 March 1983 to 29 February 1984

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SUMMARY

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Further oxidation studies of the n-alkylated aromatics has been completed and reveal insights with respect to the oxidation of the alkyl side chains. Toluene is unique in its oxidation due to the nonreactivity of the benzyl radical that forms. The higher normal alkyl aromatics all form styrene and allyl radicals during the initial stages of oxidation. Thus the oxidation of these fuels will all be similar to ethyl benzene. The first results on the oxidation of 1-methyl naphthalene have been obtained. A study on the oxidation and pyrolysis of butadiene, an important aromatic intermediate and soot precursor, has been completed. More extensive experimentation on the sooting tendencies in premixed and diffusion flames has been performed. The new fuels used were the cycloalkanes, the cycloolefins, purified 1,3-butadiene and 1-methyl naphthalene. A new model for sooting under premixed conditions reveals that original fuel structure does not play a role in soot production, but the number of carbon-carbon bonds in the fuel do. Diffusion flame results confirm that fuel pyrolysis rates and intermediates determine the sooting tendency in this combustion system. The new theory of boron ignition and combustion has been improved further. Appropriate droplet generators for boron and carbon slurries have been developed.

I. INTRODUCTION

Interest in improving performance/emission characteristics of propulsion systems requires better understanding of the coupling between fluid mechanical and chemical kinetic processes which occur in these devices. Modelling of the necessary combustion chemistry is impeded by the lack of adequate knowledge as to the general and detailed features of the kinetic mechanisms. More recently, uncertainties in the future sources and characteristics of fuels has emphasized the need to better understand fuel effects on combustion, e.g. energy release distribution, soot formation, emissions, etc. Such information is particularly lacking for aromatic species which now are comprising a major fraction of present fuels and could be a dominant factor in future jet propulsion fuels. Recent interest in high performance, high density fuels and propellants such as boron and carbon slurries has made it evident that the combustion characteristics of these materials must be understood before their eventual potential can be reached in use. Thus research endeavors at Princeton under the subject program include pyrolysis and oxidation of aliphatic hydrocarbons, pyrolysis and oxidation of aromatics, mechanistic studies of soot formation/destruction, and boron and carbon slurry combustion in turbulent reacting flows.

The specific objectives of the effort have been:

1. To initiate quantitative studies on the pyrolysis and oxidation of aromatics.
2. To continue studies of the pyrolysis reactions of the higher aliphatic hydrocarbons with and without the presence of small concentrations of oxygen.
3. To study soot formation and destruction processes in premixed and diffusion limited systems.
4. To initiate work on boron and carbon slurries, both on a particle and cloud basis, and in consideration with turbulent reacting flow.

II. STATUS OF RESEARCH

A. Introduction

In the following sections the principal tasks and results of the program are discussed.

1. aromatic pyrolysis and oxidation
2. aliphatic hydrocarbon pyrolysis kinetics in the presence of various oxygen concentrations
3. soot formation and destruction mechanisms
4. boron and carbon slurry combustion in turbulent reacting flows

B. The Oxidation and Pyrolysis of Aromatic Hydrocarbons

An initial approach of the fuels research program was to challenge what most thought to be a most complex area of chemical kinetics, that is, the oxidation processes of the aromatic hydrocarbons. The approach used was to survey and analyze the literature in order to guide the experimentation. The motivation was that again the unique characteristics of the Princeton turbulent flow reactor would permit insights into this complex system. This flow reactor shown in Figure 1 has been described in detail in various publications [1,2,3]. Most of the very early published work on aromatics was performed at low temperatures, nevertheless, proceeding to higher temperatures closer to combustion process levels was thought of as possibly clarifying the mechanism. High temperature mechanisms are needed. The analysis of the low temperature work created enough outside interest to warrant publication [4] and led to the questioning of the applicability at the higher temperatures of the earlier mechanism proposed.

Specifically, the objectives of this aspect of the program have been to elucidate the mechanisms of the oxidation of aromatic fuels with a particular

emphasis on those aspects of the mechanisms which lead to soot and other pollutant formation, and to develop, where possible, specific chemical kinetic data on the individual reactions which make up the complex reaction history of the aromatic hydrocarbons. These results would then permit the development of overall rate expressions for combustor modellers, as had been so successfully done for the alkyl hydrocarbons [5].

As previously reported [6], a prominent factor in understanding aromatic oxidation has been the actual understanding of how the phenyl radical was oxidized. Extensive data and a proposed mechanistic explanation have been published [6,7]. During this year, the effort concentrated on the oxidation of the side chain in alkylated aromatics and in obtaining some preliminary results on 1-methyl naphthalene. These compounds, and particularly the polynuclear aromatic 1-methyl naphthalene, form the major aromatic components of jet propulsion fuels.

Much has been accomplished in these studies of the alkylated aromatics. Since the interest is in the attack of the alkyl group which is the side chain attacked early in the oxidation process, the flow reactor was operated at higher velocities and under conditions so that the early stages of the reaction were spread over a larger region and thus more readily analyzed. Much of this work has been written for publication [8,9,10].

Shown in Figures 2 and 3 are results for some toluene oxidation experiments performed under the spread conditions. Particularly to be noted is the rather large concentrations of dibenzyl ($C_{14}H_{14}$) which formed. This result indicated that the benzyl radical was not very reactive, particularly with O_2 , and that indeed it probably could only be oxidized by the radicals O and OH . The similarity to the situation with respect to the methyl radical immediately became evident. With this conceptual idea, the experimentation and analysis proceeded. The very interesting results are now summarized [9].

Though the benzyl radical is formed very easily in the initial stages of toluene oxidation and some of these radicals become the dimer dibenzyl, the absence of stilbene under both rich and lean conditions suggests that dibenzyl formation does not provide an alternative degradation path as ethane formation does during methane oxidation [1]. However the experiments showed most surprisingly that even under fuel rich conditions, the methyl side chain experienced essentially a fuel lean environment. Perhaps at an extremely rich condition one

in which even all the available oxygen would not be sufficient to consume just the methyl side chain, a decomposition involving dibenzyl could evolve.

With these toluene results and the ethyl benzene results [10] to be discussed later, it was concluded that toluene oxidation may be as unrepresentative of the oxidation of longer chain n-alkylated aromatics at flow reactor temperatures as methane oxidation is unrepresentative of the general mechanism of alkane oxidation.

From a very fundamental point of view, it had been found that, at temperatures around 1000 K and at atmospheric pressure, the reactions of benzyl radicals with radicals are unusually important in the formation of intermediate species. The association reactions of benzyl radicals with hydroperoxyl radicals and oxygen atoms are the primary sources of benzaldehyde formed. The association reaction of benzyl radical with OH was responsible for the benzyl alcohol formed. At higher temperatures the significance of these reactions could change somewhat, because of the decrease in the hydroperoxyl radicals. But higher temperatures increase O and OH concentrations and the importance of these associations become greater.

Figures 4 and 5 show some of the new data on the oxidation of ethylbenzene. The results indicate that the ethylbenzene is rapidly converted to styrene through abstraction of a benzylic hydrogen by a hydroxyl radical and then the loss of a second hydrogen by O₂ abstraction or a pyrolytic step. After its formation, the styrene may undergo a hydrogen displacement reaction to produce benzene and a vinyl radical, or it may be attacked by atomic oxygen at the terminal carbon to produce a benzyl radical. This benzyl radical will oxidize to yield benzaldehyde which subsequently reacts to yield a phenyl radical. Of course, much the same happens to the benzyl radical in the toluene oxidation discussed. It again must be emphasized that these results hold for the temperature range of this study. Pyrolytic cleavage of the sidechain could be much more important at flame temperatures than as found in these studies.

The conversion of ethylbenzene to styrene and the oxidative attack on the styrene are quite analogous to reactions which occur in the oxidation of ethane. Indeed the potential analogy between the early reactions of the alkylated aromatics and those of the alkanes is utilized to predict that, after the abstraction of a benzylic hydrogen, the n-alkylated aromatics above ethylbenzene will undergo α -scission to form styrene and an alkyl radical. Initial flow reactor

data on the oxidation of propylbenzene at 1060 K show that these reactions to form styrene represent the major fuel consumption path. Thus it begins to appear that one can propose for the n-alkylated aromatics a generalized mechanism which consists of reactions to remove the side chain and those involved in the oxidation of the phenyl radical. This mechanism is the first step in the eventual goal of an overall mechanism for aromatics to be provided to modelers.

The initial results on the oxidation of 1-methyl naphthalene were presented at an A.F.O.S.R. Contractors' Meeting. Since these results were the first taken, no extensive discussion will be given here. What is important is that these results show that the experimentation and analyses equipment were capable of studying this important polynuclear aromatic which is a component of jet propulsion fuels.

C. Pyrolysis and Oxidation of Aliphatic Hydrocarbons

The initial motivation behind this element of the total effort was to provide basic rate data and mechanisms to an element of the fuel combustion process not studied as extensively as the others. Fuel pyrolysis studies and the effect of small concentration of oxygen on fuel pyrolysis were considered essential to the understanding of the burning processes in fuel-rich pockets. That this concept was an important and necessary element of the combustion system was vindicated, it is thought, by the results obtained in the aromatic oxidation and soot formation efforts of this program. As will be discussed, fuel pyrolysis is found to be the essential controlling factor in soot formation in diffusion flames. In this sense the term pyrolysis refers to the pyrolysis reaction rate and character of the intermediates formed. Last year extensive results [11] were reported on the pyrolysis of aliphatics with and without addition of small concentrations of oxygen. The results of this phase of this effort were very important in analyzing [12] the possible different effects of oxygen in sooting diffusing flames when one is comparing aliphatics and aromatics. Both with respect to understanding further fuel structure effects in sooting diffusion flames and the fate of the important intermediates (such as butadiene and vinyl acetylene) in aromatic oxidation, this phase of our work was continued during the present contract period by a detailed study of butadiene pyrolysis and oxidation.

As indicated, progress in this phase of the effort was made with respect to the pyrolysis and oxidative mechanisms of butadiene, an important compound with

respect to soot precursors and aromatic intermediates. Again, since the results of this work were recently prepared for the open literature [13,14], only a summary will be presented.

There are many intermediates found in the oxidation and pyrolysis of 1,3-butadiene. Indeed these intermediates permit one to understand the many reactions which take place. Instead, for brevity, the only figure to be presented in this annual report is that which shows how the butadiene decays as the initial oxygen concentration is varied. Shown in Figure 6 is the rate of butadiene disappearance as a function of initial equivalence ratio. The data represent a variation from a very lean run ($\phi = 0.55$) to a pure pyrolysis run ($\phi = \infty$).

A mechanism has been postulated [13,14] and is based on the nature of the intermediate species observed, their relative order of maximum concentration and some assumptions about high temperature reactions of radicals with olefins. The main points of the mechanism are:

1. Butadiene decay in the presence of oxygen appears to be the result of a true oxidative attack rather than some oxidative abstraction that would make oxygen appear as a catalyst to early pyrolysis.
2. The oxidation of butadiene appears to occur through O atom addition to the double bond. Abstraction reactions involving OH and H to form the important butadienyl radical, C_4H_5 , also play some role.
3. The formation of acrolein appears to result from the reaction of allyl radical with either O or HO_2 .
4. Many routes lead to the formation of copious amounts of ethene and acetylene.

D. Soot Formation and Destruction Processes

1. Introduction

The major objectives of the soot studies have been:

- a. a complete and critical review of the soot literature in order to evolve phenomenological models and understanding of the overall processes so that control strategies could be developed.
- b. experimental investigations with gaseous fuel jets (laminar diffusion flames) in order to evaluate the effects of water and other additives on the soot formation process and to scrutinize the models developed.
- c. experimental investigations with premixed laminar flames for the same purposes as (b).

- d. the development of a rapid experimental test to permit evaluation of the sooting tendency of practical liquid fuels, particularly those containing fractions of aromatics.

Items (a) and (d) have been completed and were reported as part of five major publications [15-19]. The phenomenological models and the results obtained under Items (b) and (c) have given substantial understanding of soot processes, much of which has practical application. Indeed, this A.F.O.S.R. work has had substantial influence on the direction and thinking of many investigators in the soot field.

2. Research Progress

a. General Background

The review and phenomenological models developed have been published as an A.F.O.S.R. report [15]. The extensive information reported, will not be repeated, but the major conceptual ideas promulgated will be reviewed.

The complete soot mechanism in a combustion system is truly very complex. The process may be described in a cursory fashion as follows. Whether starting in a premixed or diffusion flame system, a fuel undergoes pyrolysis and eventually forms a species which is the precursor to the actual soot nucleation process. The precursor then reacts or polymerizes in some manner so that soot nuclei form. During this process it is feasible that some of the precursors (or even the species forming the precursors) can be oxidized. Nuclei which form can absorb other high molecular weight hydrocarbons which exist in the combustion system. Since the particles exist in high temperature regions, they can undergo condensed phase reactions which can reduce their hydrogen content and eventually give the highly complex aromatic structure we know as soot. The soot particles can conglomerate and agglomerate while these processes are proceeding. Simultaneously, there can also be oxidation of the particles.

Although the overall soot formation process as defined above seems very complex, the Princeton research has been governed by the essential idea that the controlling factors in most sooting combustion systems are the rate of formation of the precursors and the rate of oxidation of the formed particulates.

Most investigators have been concentrating their efforts on the nucleation process. Although nucleation is obviously an important step, recent results obtained seem to confirm that it is not the controlling one. Nevertheless, the initial review focused on this question and it was considerations which grew out

of this study that guided much of our original experimentation on sooting diffusion flames.

The two primary mechanisms that have been suggested in the past have been the polyacetylene route and ion-neutral route. Doubts have been raised about the polyacetylene route because it is known that in flame processes soot forms very early. The time required for the numerous free radical-neutral acetylene reactions, the energy required for cyclization, etc. have many to question this type of mechanism. There has been substantial evidence that polyacetylenes form in flames, but their fate may simply be to be destroyed or absorbed on particles already formed. The concept of fast ion-neutral reactions being the nucleation route is also open to serious question. Firstly, there are much smaller ion concentrations in diffusion flames than in premixed flames, yet the propensity to soot in diffusion flames is far greater than in premixed flames. More importantly, the ion concentrations in flames are orders of magnitudes lower than the formed particle density. Thus it is quite possible that there simply are not sufficient ions present to nucleate all particles.

A suggestion of Thomas [21] that Diels-Alder (condensation) type reactions were fast and a possible nucleation route for soot formation directed the original Princeton conceptions. The thinking followed the lines that any soot precursor must be highly conjugated to be stable at the high temperatures in flames, and, indeed the high molecular weight species forming from these precursors must be conjugated as well. Conceptually then one could understand why the aromatics had a strong tendency to soot. In a very classical sense, Diels-Alder reactions are very much like electrostatic reactions and if one examines the resonance structures of most compounds which undergo Diels-Alder reaction, one can understand from the polar nature of some of these structures the possibilities of these reactions. If this conceptual idea was correct, then certain compounds such as butadiene and vinyl acetylene should have a great propensity to soot. Indeed, Schalla, et al. [22] reported that butadiene had a greater tendency to soot than the aromatics. Early experimental work at Princeton had shown butadiene to be a more prolific sooter than other fuels, but not to the extent that Schalla had found. However recent results [23] with purified butadiene now make the results with this important fuel consistent with others. The evidence being obtained appears to support the contention that vinyl acetylene and butadiene, or radicals formed from these species, may be the main precursors

to soot nucleation. The fact that vinyl acetylene undergoes Diels-Alder addition to form the aromatic molecule styrene leads one to believe that it is the more important of the two precursors. Indeed the importance of the Princeton kinetic studies on the oxidation of aromatics, reported earlier [6,7], is now most evident. The fact that vinyl acetylene and butadiene form in the oxidation of aromatics may be the key to understanding soot formation processes with aromatic fuels. However, recent estimates of the A factor in the rate expression indicate that simple Diels-Alder reactions may be too slow for the soot forming process. There is also a possibility that the butadienyl radical could readily become ionized whereby its dissociation products would also be so, and very fast Diels-Alder reactions could proceed.

Most of the early work on soot formation in premixed flames (Street and Thomas [24]) compared the relative tendency of fuels to soot and reported the tendency as:

aromatics > alcohols > paraffins > olefins > acetylenes

Initial results obtained in this program indicated that except for the aromatics, the trend was not just a matter of a homologous series trend, but the greater the flame temperature the less the tendency to soot. This conclusion was consistent with what Milliken [25] had reported from his considerations of premixed ethylene-air flames that the tendency to soot was a function of the flame temperature. Milliken calculated that the rate of "pyrolysis" of acetylene to form the soot precursors rose more slowly with temperature than the rate of oxidative attack on the precursor forming system. Thus under premixed conditions, the only reason that acetylene seems to have little tendency to soot compared to other fuels is its high flame temperature compared to the other alkyls. Since the Princeton work on the oxidation kinetics of alkyls indicated that under rich conditions all compounds break down to acetylene, it was postulated that to a first order the sooting tendency of the alkyl compounds under premixed conditions are independent of fuel structure and primarily a function of the temperature of the combustion system. It was this conceptual idea that guided the experimental work on premixed flames to be discussed subsequently.

The question of how the aromatics behave with respect to sooting is apparently a different one. Benzene, for example, has a temperature close to ethene, yet, its tendency to soot is greater. From an initial review of aromatic oxidation [4], the conclusion was reached that the mechanism was such that

carboxyl structures formed and these led to radicals which eventually formed CO_2 directly. Thus, the demands of two carbon atoms in the ring was for two oxygen atoms each compared to the fact that all alkyl compounds eventually take a route through a formyl group and only form CO . Thus it was stated that benzene at a given equivalence ratio was richer than an alkyl compound of the same C/H ratio. This condition was thought to give the great tendency of benzene to soot and not to have correspondence with respect to the temperature trend of the alkyl compounds. Again, the experimental results on the oxidation of the aromatics has helped clarify this point. The results showed that it is not likely carboxyl structures will form during the oxidation of the aromatics, certainly not at the high temperatures of combustion systems. However, the analyses also show that vinyl acetylene and butadiene form as well as acetylene. An extension of this concept is that acetylene must form such compounds to undergo soot nucleation steps. Milliken's concept was that acetylene pyrolysis to soot versus rate of oxidative attack determined the trend to soot with respect to temperature. His concept apparently would hold for the alkyl compounds because they follow the same degradation route to acetylene. The aromatics do not follow this route. Instead they go directly to important soot precursors. Thus the competitive trend with respect to precursor formation and oxidative attack could be altered substantially. However, it has been subsequently learned that under premixed conditions that all compounds, alkyl and aromatic, break down and soot forms from the acetylene formed and in the post flame zone.

The reasoning on the temperature effect under premixed conditions can be extended, quite simply, to diffusion flame systems in order to determine the temperature effect. Since there is no oxidative attack on the soot precursor process in diffusion flames, then the pyrolysis rate increase with temperature is not counteracted by any other effect. Thus the higher the fuel stoichiometric flame temperature the greater the tendency to soot. The general tendency as reported by Schalla et al. [22] long ago shows the trend according to homologous series:

aromatic > acetylenes > olefins > paraffins > alcohols

Again, except for the aromatics, the trend is the greater the stoichiometric flame temperature the greater the tendency to soot. However Schalla et al. results show that the C_4 olefins had the greatest tendency to soot among the olefins. Further branched chain isomers had a greatest tendency to soot than

the corresponding straight chain compounds. Again the characteristic of benzene pyrolysis would have to be different from the alkanes. These facts led to the conclusion that the tendency to soot in diffusion controlled systems was dominated by the flame temperature, how this temperature affects the pyrolysis zone, the initial fuel structure and the fuel pyrolysis intermediates. Indeed this concept guided much of the experimentation with fuel jet diffusion flames to be discussed next.

b. Sooting Tendency in Diffusion Flames

In earlier work it had been shown that addition of an inert did not alter the luminous flame height of a diffusion controlled system [25]. Thus by the addition of an inert to the gasified fuel one could control the temperature of a fuel diffusion flame independently. The actual measurements proceed by measuring the diffusion flame smoke heights (and the mass flow rate at this height) with various amounts of a nitrogen diluent. Since the model of soot formation was that fuel pyrolysis kinetics control, the data were plotted as the $\log (1/\text{fuel mass flow rate (FFM)})$ vs $1/T$ and straight lines were obtained for all fuels tested. In a major publication [26] results on acetylene, ethene, butane, propene, n-butene, pentene and hexane were published. Data on the xylenes, ethyl benzene and 1,3 and 1,4 cyclohexadiene had been obtained as well. During this year additional data have been obtained on the following fuels: C_3 , C_5 , and C_6 cycloolefins, purified 1,3-butadiene, C_5 and C_6 cycloolefins, and 1-methyl naphthalene.

The data on these new fuels have been reported in three recent publications [12,23,27]. Figure 7 shows a plot of $\ln (1/\text{FFM})$ vs $1/T$ for all measurements to date (current and past periods). Again, without going into great detail about each fuel, the essential characteristic that was found was that in diffusion flames two factors control the extent of soot formation, the flame temperature and the fuel pyrolysis. In essence fuel pyrolysis means initial fuel structure which determines the rate of fuel pyrolysis and the crucial intermediates which form. What is most important is that we can explain the position of any fuel shown in Figure 7, from what is known about that fuel in pure pyrolysis [12,28]. Essentially this statement means that if one knows the pyrolysis kinetics of a fuel one can predict its relative tendency to soot.

Concisely from Figure 7 then, all aromatics have relatively the same tendency to soot and have a mild temperature dependency. The aromatics, and those

fuels which pyrolyze into aromatics such as the cyclohexadienes and cyclopentene, have the greatest tendency to soot of all fuels measured. The diolefin, butadiene, is the next greatest sooter followed by the olefins and those fuels which pyrolyze to the olefins. All olefins have a much greater temperature dependency. Then comes acetylene and the paraffin. As would be expected acetylene has a very large temperature dependency. Application of these type of results to practical power plants has been discussed in a recent A.F.O.S.R. technical report [29].

c. Sooting Tendency in Premixed Flames

The qualitative procedure for evaluating sooting tendency of fuels under premixed combustion conditions has been to observe the C/O ratio (or equivalence ratio ϕ) at which luminosity just begins in laminar Bunsen type flames. The larger the C/O or equivalence ratio at which luminosity begins the less the tendency to soot. The most extensively published data of this type have been those of Street and Thomas [24]. Milliken's interpretation [25] of the competing phenomena in premixed flames made it evident that the C/O ratio was not necessarily the primary variable for categorizing fuels with respect to their tendency to soot under premixed conditions. Indeed from oxidation kinetics one would expect acetylene to soot the most, not the least, among the alkyls. Thus the experiments of the Street and Thomas type were repeated with temperature control of the flame at each mixture ratio. The control was achieved by varying the nitrogen concentration in a nitrogen-oxygen mixture. Thus for a given fuel, a temperature higher or lower than the value for air could be obtained.

An interesting experimental procedure developed in the laboratory permitted runs under very fuel rich conditions with great dilution and without stability or cellular flame problems [30]. Essentially oxygen enriched the stabilization point of a Bunsen flame. Further, it was also found the data taken on many fuels gave the most meaningful trends (i.e. with what one knows with respect to fuel pyrolysis kinetics) when plotted as the equivalence ratio (ψ) based on stoichiometry going to carbon monoxide and water vapor versus the calculated adiabatic flame temperature [30]. The fuels considered at that time were: ethane, ethene, acetylene, propane, propene, butane, 1-butene, 2-methyl propane, 2-methyl propene and 1,3-butadiene. During this year work has been completed on: hexane, 1-hexene, cyclohexane, cyclohexene, 1-octene, benzene, toluene, cumene and 1-methyl naphthalene. The critical sooting equivalence ratio ψ_c for

all these data are plotted in Figure 8.

Due to a model developed which correlates the sooting tendencies of all fuels tested in this program, it is believed that sooting tendencies under premixed conditions is nearly completely understood.

The reasoning [31,32] is as follows. Since the pyrolysis and oxidation rates follow Arrhenius kinetics, then a plot of $\log \psi_c$ versus $(1/T_f)$ should produce a straight line for each fuel. Figure 9 shows that the data explicitly correlate in this manner over the entire temperature range investigated experimentally. The natural tendency is to assume that the Arrhenius plot represents an activation energy of some controlling process in the system. Under premixed conditions, however, the inference made is that the balance between the fuel pyrolysis rate and the oxidative attack on the precursors is the controlling factor in determining the sooting tendency. Both the pyrolysis and oxidation attack follow Arrhenius kinetics and thus the method of plotting should give straight lines for the data. One should notice that the slope of each fuel line is almost identical. The apparent activation energies determined from Figure 9 are within the range of 3-6 kcal/mol for all fuels examined.

In order to understand the effects of the flame temperature, the number of carbon atoms, the C/H ratio, and the fuel structure, an overall analysis of the soot formation process would be useful. Millikan's [25] formulation of the problem may be modified by considering the initial fuel generally instead of acetylene explicitly. Consequently the rate of soot precursor formation may be taken as zero at the critical mixture ratio at which soot just appears; i.e.,

$$\frac{d[P]}{dt} = k_p [\text{Fuel}] - k_o [\text{OH}] [P]_c = 0. \quad (1)$$

where $[P]_c$ is the critical concentration of precursors that gives soot.

By using Arrhenius parameters, the OH concentration then may be written as

$$\log [\text{OH}]_c = \frac{E_o - E_p}{2.3 RT} + \log \frac{A_p}{A_o} + \log [\text{Fuel}]_c - \log [P]_c \quad (2)$$

where the subscript c expresses the critical condition. The equilibrium OH concentration may be utilized in order to observe the differences due to fuel type. The equilibrium OH concentration may be written as

$$\log [\text{OH}]_{eq} = a \frac{1}{T} + b + c \log \psi. \quad (3)$$

The fuel concentration in the mixture is an approximately linear function of ψ in the range of the fuel mole fraction less than 0.1. Then, at the critical condition,

$$[\text{Fuel}]_c = d \psi_c . \quad (4)$$

By substituting Eqs. (3) and (4) into Eq. (2) for the critical condition, we find that

$$(1-c) \log \psi_c = - \frac{E_o - E_p}{2.3 R} + a \frac{1}{T} + b - \log \frac{A_p}{A_o} - \log d + \log [P]_c \quad (5)$$

or

$$\log \psi_c = - \frac{\frac{E_o - E_p}{2.3 R} + a}{(1-c)} \cdot \frac{1}{T} + C , \quad (6)$$

where C contains all terms independent of T.

Thus, the temperature effect on the sooting limit may be evaluated, in an overall sense, in terms of the activation energies of the fuel pyrolysis (E_p) and the precursor oxidation process (E_o) and the dependencies of the OH radical concentration on the flame temperatures (a) and the effective equivalence ratio (c). Fuel pyrolysis studies [11] have revealed that the overall activation energies of pure pyrolyses are approximately the same (65 kcal/mol) for six aliphatic fuels (propane, butane, isobutane, 1-butene, hexane, and 1-hexene). The activation energies for the OH radical attack on the soot precursors also do not vary greatly with initial fuel type and must be very small, probably 0-7 kcal/mol. The equilibrium OH concentration for several fuel/oxygen/nitrogen mixtures at $\psi = 1$ was calculated. The most significant feature as would be expected, is that the slope of each fuel OH line as a function of ($1/T$) is identical for all fuels ($a = -1.6 \times 10^4$). The apparent "activation energy" determined from the slope is 74 kcal/mol. The coefficient c is about -2.5 for all fuels in Figure 9 except for benzene (-3.1) and acetylene (-3.3). Thus almost all factors which determine the temperature effect on the sooting tendency are minimally related to the fuel type. The sum of the apparent "activation energy" of the OH radical concentration and the activation energy of the precursor oxidation process as calculated is somewhat larger than the estimated activation energy of the fuel pyrolysis process. The arguments made would dictate that the difference in these two values should not change with the fuel type. It is rather remarkable that the experimental data as correlated in Figure 9 not only

reveal this fact, but also show a slope (3-6 kcal/mole) that is almost the same as that estimated (1-3 kcal/mol) from Equation (6).

The coefficient b or the OH concentration at a fixed T_f and ψ , decreases monotonically with increasing C/H ratio and does not depend on either the number of fuel carbon atoms or the structure. Thus, increasing the C/H ratio at a given number of carbon atoms reduces the oxidation rate and increases the tendency to soot. On the other hand, for a homologous series the overall reaction rate constant for pure pyrolysis at a fixed temperature (or A_p) increases dramatically with increasing number of carbon atoms [11]. The coefficient d decreases with increasing carbon number, but this effect would be less important as compared with the pyrolysis rate effect. Therefore, the increase in the sooting tendency with increased number of the carbon atoms can be explained by the increase in the fuel pyrolysis rate.

Figure 10 shows the heuristic correlation between the critical effective equivalence ratio to soot (ψ_c) at a fixed flame temperature (2200 K) and the "number of C-C bonds" in which a double bond is counted as two and a triple bond as three. The total C-C bond strength which almost linearly depends on the "number of C-C bonds" can be used as well [28,30]. All data, even those for the aromatics, lie on a single curve. Thus it appears possible to categorize fuels with respect to their degree of sooting under premixed conditions by use of a single property of the fuel, the "number of C-C bonds" or the total C-C bond strength. This correlation has further significance [29] in that it reveals that aromatic structure is not significant under premixed sooting conditions and leads us to propose that in this type of combustion system all fuels break down to elemental species which then build up to the precursors. Obviously molecular size has an effect.

E. Boron and Carbon Slurry Combustion in Turbulent Reacting Flows

1. Progress in Slurry Work

Advances have been made in the work on slurry droplet combustion and on turbulent flame theory. In addition, a paper [33] on boron particle combustion has been submitted to the Twentieth International Symposium on Combustion and is essentially an extension of earlier work [34]. The status and progress of the work in each of these areas is summarized below.

a. Slurry Droplet Combustion

The work on slurries is directed toward obtaining a fundamental

understanding mechanisms of combustion of single slurry droplets. This understanding is needed to provide direction for investigation of useful modifications of combustion systems and fuel formulation to improve the performance of slurry-fueled propulsion devices.

Questions had been raised concerning the influence of support fibers on the combustion behavior of fiber suspended slurry droplets. Further consideration of these difficulties, which appear to be especially severe on the aspects which are of particular interest here (e.g. agglomeration of slurry particles, achievement of disruptive burning, etc.) have prompted the investment of a substantial effort in developing a suitable technique for producing monodisperse, isolated free droplets of slurry fuels which could then be projected into well defined combustion environments. During his sabbatical (1982-83 academic year), one of the principal investigators (F.D.) thoroughly reviewed and experimentally investigated several droplet generation techniques, including the Rayleigh instability method, the capillary jet chopping method, and the piezoelectric ink-jet printing technique and found that each of these methods was limited to the use of very dilute slurries or the production of initial drop sizes large than 800 to 1000 microns, and/or could not produce droplets with sufficient spacing. He and his colleagues at Mobil Research Corporation's Central Research Division initiated development of a droplet generator technique which has the potential to overcome these difficulties.

The device basically consists of a capillary tube through which a liquid flows, with the tube exit positioned co-linearly along the axis of a high velocity nitrogen gas jet. A stream of droplets is produced by inducing the detachment of the droplet that is formed at the end of the capillary jet by surface and interfacial tension effects. By varying the capillary liquid flow rate, the nitrogen gas purge velocity and the capillary size, a stream of varying droplet size, spacing and initial droplet velocity can be precisely controlled. Initial work was performed using a 32 gauge (100 micron diameter) capillary tube positioned along the axis of a venturi with a minimum inside diameter of 1000 microns. The exit of the capillary was positioned very near the vena-contracta of the venturi flow (see Figure 11). Using hexadecane as a liquid test fuel, droplet diameters in the 250-425 micron range with spacings of 100 to 650 droplet diameters and initial velocities of 2-3 meters per second could be produced with as little fuel flow as 8 ml/hr. Similar results were easily

established with higher viscosity liquids such as marine residual fuel oil. The technique was simpler than those mentioned earlier, both in required hardware and in mechanism of achieving droplet isolation. Furthermore, the technique required substantially less fuel flow and successfully operated on higher viscosity materials. However, initial tests with 10% boron slurry in JP-10 fuel were discouraging. The small capillary diameter caused separation of the slurry particles from the JP-10 at the inlet to the capillary and eventual clogging of the system. Significant progress in alleviation of this difficulty has been made, and completion of a droplet generation scheme which will successfully operate on slurry fuel is near. The primary improvements have been as follows.

i. Liquid Fuel Supply System

A fine steel wire is inserted through the hypodermic capillary tubing (Figure 12) so that droplets are formed and removed at the tip of the wire rather than at the capillary exit itself. Thus it is possible to produce droplets with initial diameters near the diameter of the fine wire. By replacing the the wire with ones of various size a large range of initial droplet size can be achieved. But the more important advantage is that a hypodermic inside diameter large enough to alleviate clogging by the slurry can be accomodated without affecting the droplet size produced.

ii. Gas Purge Flow Configuration

The venturi type flow system has been replaced with a simple converging nozzle (Figure 13). This has resulted in a significant reduction in the scatter of the trajectories follwed by the droplets formed.

An initial investigation of these modifications using hexadecane has been completed and it has been found that it is possible to produce initial droplet sizes as small as 100 microns with capillary inside diameters as large as 500 microns.

In the course of this work, a simple analysis of the droplet motion to obtain information on parameters controlling initial droplet velocity also has been performed. The original intent of this work was to investigate whether the technique could be successfully employed in a vertically upward oriented as well as a vertically downward oriented experiment. The droplet was simulated as a solid sphere initially at rest, accelerated by both gravity and drag forces after detachment. Thus, the forces acting on the sphere are;

$$F = - \dot{m}_D + \pi r_D^2 \cdot 1/2 \rho_G (v_G - v_D)^2 \quad f \quad (1)$$

where

$$f = \frac{24}{\frac{2r_D (v_G - v_D) \rho_G}{\mu_G}} = \frac{24}{Re_D} \quad (Re_D < 2) \quad (2)$$

$$f = \frac{18.5}{Re_D^{3/5}} \quad (2 < Re_D < 5 \times 10^2) \quad (3)$$

$$f = .44 \quad (5 \times 10^2 < Re_D < 2 \times 10^5) \quad (4)$$

The subscripts D and G denote droplet and gas, respectively. The symbols m, r, rho, v, f, mu, and Re represent mass, radius, density, velocity, drag coefficient, coefficient of viscosity, and Reynold's number respectively. Thus, the force is set equal to the rate of change in the droplet momentum, i.e.

$$\dot{m}_D v_D = - \dot{m}_D + \pi r_D^2 \cdot 1/2 \rho_G (v_G - v_D)^2 \cdot f \quad (5)$$

where the dot expresses the time derivative. This equation can be solved analytically for Stokes' flow ($Re < 2$), i.e.

$$v_D = - \frac{Q}{P} (1 - e^{-Pt}) \quad (6)$$

$$x_D = - \frac{Q}{P} (Pt + e^{-Pt} - 1) \quad (7)$$

$$\text{where } P = \frac{G \pi \mu_G r_D}{m}, \quad Q = g - P v_B \quad (8)$$

and the initial conditions are:

$$v_D = x_D = 0 \quad \text{at } t = 0$$

Although it is necessary to solve the equation numerically for higher Reynold's numbers, it is possible for a first approximation to obtain a solution by multiplying the Stokes' flow solution by the ratio of the drag coefficients given by Equations 3 and 4 to those yielded by Equation 2. Figure 14 shows the relative magnitudes of the drag and gravitational forces acting on the droplet. These results were calculated using the experimental measurements taken on the first prototype system with vertically downward droplet generation. It is clear that the drag force is the determining parameter. Figure 15 shows the acceleration of the droplets in constant velocity gas flow, calculated by Equations 6 and 7. The droplets are seen to be accelerated to velocities observed in the experiments in less than 1 cm distance of travel.

These data have been useful in designing the generator now beginning to be tested with slurries. The data of Figure 14 also suggest that the droplet generator technique should be easily adapted to a vertically upward oriented droplet combustion experiment such as those typically performed here at Princeton. However, in further experiments it has been found that the surface tension effects which occur near the tip of the capillary cause the fuel to flow down the outside of the capillary in the vertically upward orientation, and the liquid layer only sporadically extends far enough into the gas flow to result in detachment. Droplets of variable size and frequency of detachment result. The experimental droplet combustion facility to accommodate running the experiment in a vertically downward orientation is therefore presently being redesigned. An experimental facility of this design was designed and constructed elsewhere by Professor Dryer during his sabbatical, and results of that work are guiding the current effort. These modifications are expected to be complete and testing of the new droplet generator to proceed such that initial data on slurry droplet combustion will be forthcoming soon.

In addition to this experimental work, appreciable progress has been made in the research on the theory of slurry droplet combustion. The objectives of this work have been to define and analyze simplified models of the combustion of slurry droplets. The models then provide predictions of parametric dependences of burning properties on fuel properties and on experimental conditions. These predictions can be tested experimentally to ascertain how good the models are. Obtaining a successful simplified model appreciably improves the understanding of the combustion process and enables extrapolations to conditions not investigated experimentally to be made. Unsuccessful comparisons can lead to improvements in modelling concepts.

Two simplified models for the combustion of slurry droplets were identified, a contracting-sphere model and a porous-shell model. Preliminary experimental observations of the combustion of fiber-supported slurry droplets having boron loadings in the range of practical interest indicated that the porous-shell model is the more realistic of the two. Therefore a detailed theoretical study was devoted to the porous-shell model, and the analysis thereof has now been completed. Numerical predictions for test cases currently are being performed. The character of the model and the nature of the analysis and of the results are described below.

The simplified model is one in which when the liquid phase evaporates a porous solid agglomerate is left behind. Experimentally the agglomerates are observed and, depending on conditions, contract somewhat or swell during combustion of the liquid fuel. As an approximation it was assumed that the porous agglomerate is rigid and maintains a radius equal to the initial radius of the droplet. Two stages of combustion were considered. In the first stage, heatup of the slurry sphere was analyzed with evaporation neglected in the first approximation. This stage ends when the surface temperature of the droplet reaches the equilibrium wet-bulb temperature of a burning droplet, typically only slightly below the liquid boiling temperature. In the second stage the droplet consists of a porous shell of fixed outer radius, concentric with an inner sphere of a liquid-solid mixture, whose surface regresses because of liquid vaporization (see Figure 16). The second stage persists until the liquid component is entirely vaporized. For a number of reasons no analysis is performed for what may be considered a third stage of combustion; that is, the events after liquid vaporization.

From previous work on burning of boron particles, for example, it is clear that under many conditions the third stage, agglomerate combustion, would be by far the largest of the three. In fact, in many of the Princeton laboratory tests of fiber-supported droplets the agglomerate did not burn at all. Conditions of relatively high ambient temperature and oxygen content are needed for agglomerate combustion in a reasonable time. These conditions can be estimated from theory of boron particle combustion and are found to be prohibitively long for a number of applications. In such applications it is therefore necessary that the droplet be prevented from reaching the third stage. The analyses of the first two stages are designed to enable predictions to be made of conditions under which disruption will occur, so that the third stage will not be encountered. Thus, although the first two stages typically might extend over only a small part of the total burning time, their analysis appears to be of critical importance.

Prediction of droplet disruption depends strongly on temperature and composition profiles within the droplet. The analyses are directed toward calculating these profiles. Disruption aspects of the combustion have not yet been addressed. There are theories in the literature that have considered this question, although not specifically for slurry droplets, and further advances in

these theories are underway at Princeton in connection with another program. The results reported here concern only the combustion histories in the absence of disruption. The intent is to use these results in further studies to predict disruption as a consequence of addition of a second liquid fuel or of an energetic constituent.

A number of specific simplifying assumptions were made in the slurry-droplet burning analyses. In particular, the gas-phase processes always were treated as quasisteady. The full set of assumptions will not be listed here. For the first stage, the surface temperature of the droplet was approximated as $T_{\infty} + (T_{\infty} - T_0) e^{-ct}$, where T_0 is the initial temperature, T_{∞} an effective long-time surface temperature obtained from the gas-phase analysis and c a constant reciprocal time, also obtained from the gas-phase analysis, now coupled with the condensed-phase analysis. The condensed-phase problem involves transient heat conduction within the sphere. Solutions to the coupled problem were obtained by Laplace-transform methods and also by numerical integration of the partial differential equations. These results were used to obtain c , after which Laplace-transform methods provided a simpler form for the solution for the transient temperature profiles within the droplet. The results obtained are in good agreement with related results found earlier by other investigators.

Results for the first stage provide initial conditions for analysis of the second stage. It is assumed that during the second stage the temperature difference across the porous shell is negligibly small. Estimates for typical values of parameters have indicated that this approximation usually is reasonably good for the conditions of greatest interest. For the second stage a partial differential equation then is obtained for the temperature profile within the liquid-solid mixture and for the time rate of change of the location of the inner surface of the porous shell. This equation is solved by an asymptotic analysis for large values of the transfer number B . The result is a series expression for the time-dependent temperature profiles within the droplet and an asymptotic expansion in B^{-1} giving the time dependence of the inner radius as well as the burning time t_f , defined as the interval from the beginning of the second stage to the time of disappearance of the liquid.

A conventional "d-square" law is obtained for t_b , in that t_f is proportional to the square of the initial droplet diameter. The coefficient in this d-square law differs from that of the classical droplet-burning formula because

of the difference in the burning mechanism produced by the presence of the porous shell. The physical reasons for the difference have been investigated and understood. Concurrent with this d-square law for t_b is a d-cube law for the time variation of the diameter of the inner liquid-solid mixture; the cube of this diameter decreases linearly with time. The physical reason for this difference also is readily understandable from the model. Thought is being given to possible experimental methods for testing this last prediction in the laboratory.

b. Boron Suspension Combustion

The combustion of boron suspensions involves the burning of individual boron particles with oxidizing gases in clouds. The focus during this year has been directed strongly on the slurry combustion problem. The only progress in this area has been further refinement of earlier theoretical studies on the combustion of individual boron particles. These refinements resulted in preparation of a paper [33] on the subject, submitted to the Twentieth International Symposium on Combustion. The principal results of this theoretical works have been summarized previously. The major attention this year was to get the experimental work on slurry-droplet combustion well underway before returning to experiments on cloud combustion.

2. Progress in Turbulent Flame Theory Work

The continuing theoretical work on turbulent flame theory has resulted in two new publications [35,36], both co-authored with P.A. Libby. The focus of this work has been on ascertaining effects of variable density on the dynamics and structures of premixed laminar flamelets in turbulent flows.

In one of these studies, the method of activation energy asymptotics is used to describe the behavior and characteristics of nonadiabatic flamelets involving counterflowing reactants and products under the assumption of a unity Lewis number. For moderate and low rates of strain the results are analogous to those obtained in earlier applications of the method, namely reaction zones which are maintained in a first approximation by a diffusive-reactive balance. Indeed for low rates of strain many features of flamelet behavior are independent of the extent of the nonadiabaticity since the reaction zone is insulated from the stream of altered enthalpy by a diffusive-convective zone near the stagnation point. Two limiting processes are considered. One pertains to nearly adiabatic flamelets and exposes the principal qualitative results of this

study. The second pertains to the full, nonadiabatic case. Flamelets with product streams having elevated enthalpies are shown to possess essentially the same features as the adiabatic flamelets studied earlier. On the contrary, flamelets with cooled product streams involve complex behavior depending on the extent of the nonadiabaticity and the rate of strain. In particular the continuous behavior shown to prevail in an earlier study when adiabatic flamelets are subjected to varying rates of strain is replaced if the product stream is suitably cooled by abrupt transitions involving altered locations of the reaction zone and altered rates of creation of product. These transitions are interpreted in terms of extinction and ignition. A special treatment is provided for flamelets subjected to high rates of strain and thus involving reaction zones with diffusion, convection and chemical reaction all operative. Density variations of practical interest are considered in the numerical examples.

In the other study, the method of activation energy asymptotics is used to study the effects of Lewis numbers different from unity on nonadiabatic flamelets in counterflowing streams of reactants and products. A sequence of analyses parallels those reported earlier for such flamelets having Lewis number unity. Thus initial results relate to nearly adiabatic flows with Lewis numbers close to unity. It is found that the effect of nonunity Lewis numbers is accentuated in flamelets subjected to low rates of strain and that Lewis numbers greater than unity tend to promote extinction. Thus abrupt extinction and ignition events can occur even under adiabatic conditions. Next fully nonadiabatic flamelets with Lewis numbers near unity are treated in order to consider cases involving relatively large degrees of product heating and cooling. These results relate to reaction zones as they arise under conditions of low-to-moderate rates of strain with the customary diffusive-reactive balance. Flamelets subjected to such high rates of strain that the reaction zone is extended and located far into the product stream are also treated. In this case a diffusive-convective-reactive balance prevails. Realistic density variations are considered in the numerical examples and are shown to tend to retard extinction.

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T.A. Litzinger, K. Brezinsky and I. Glassman, "On the Oxidation of Ethylbenzene and Higher Alkylated Aromatics", to be submitted to Comb. Sci. and Tech.

V. PROFESSIONAL PERSONNEL

Prof. Frederick L. Dryer, Co-Principal Investigator
Prof. Irvin Glassman, Co-Principal Investigator, Correspondent
Prof. Forman A. Williams, Co-Principal Investigator

Dr. Kenneth Brezinsky, Research Staff Member
Dr. Kozo Saito, Research Staff Member
Dr. Fumiaki Takahashi, Research Staff Member

Mr. Allesandro Gomez, Graduate Student
Mr. Thomas Litzinger, Graduate Student
Mr. Paul Antaki, Graduate Student
Mr. George Sidebotham, Graduate Student
Mr. Edward Burke, Graduate Student

Ms. Catherine Calillo, Undergraduate Student
Mr. Jamie Bonini, Undergraduate Student
Ms. Katherine Chesmel, Undergraduate Student

VI. DEGREES AWARDED

S.L. Murphy, "Pure and Oxidative Pyrolysis of Hydrocarbons in Relation to the Sooting Diffusion Flame", M.S.E. Thesis, Department of Mechanical and Aerospace Engineering, Princeton University, August 1982.

E.J. Burke, "A Study of the High Temperature Oxidation of 1,3-Butadiene", M.S.E. Thesis, Department of Mechanical and Aerospace Engineering, Princeton University, October 1983.

VII. INTERACTIONS

A. Spoken Papers

1. By I. Glassman

"High Temperature Oxidation of Aromatics", 19th Symposium International on Combustion, Technion, Haifa, Israel, August 10, 1982.

"Theoretical Aspects of Flame Spread in an Opposed Flow Over Flat Surfaces of Solid Fuels", 19th Symposium International on Combustion, Technion, Haifa, Israel, August 10, 1982.

"A Physical and Chemical Interpretation of Boron Particle Combustion", 19th JANNAF Combustion Meeting, NASA/Goddard Space Flight Center, Greenbelt, MD, Oct. 4, 1982.

"Advanced Fuels Combustion Research", 1982 AFOSR Contractors' Meeting on Air Breathing Combustion and Explosion Research, Clearwater Beach, FL, November 3, 1982.

"Physical and Chemical Effects on Soot Formation", American Chemical Society Annual National Meeting, Seattle, WA, March 24, 1983.

"Chemical Effects in Soot Formation", Naval Research Laboratory, Washington, D.C., April 31, 1983.

"The Oxidation of Alkylated Aromatics", Italian Section of the Combustion Institute, Sorrento, Italy, July 4, 1983.

"Soot Formation Processes and the Oxidation of Aromatics", 1983 AFOSR Contractors' Meeting on Air Breathing Combustion Dynamics Research, Scottsdale, AZ, September 19, 1983.

"Soot Formation in Pre-Mixed and Diffusion Flames", United Technologies Research Center, East Hartford, CT, October 4, 1983.

"Physical and Chemical Effects in Soot Formation", New York University, N.Y.C., NY, October 17, 1983.

"Universal Hydrocarbon-Air Temperature Graphs", Eastern States Section/The Combustion Institute Meeting, Providence, RI, November 8, 1983.

"Soot Formation", Center for Energy and Environmental Studies, Princeton University, Princeton, NJ, February 7, 1984.

"Chemical Kinetic Aspects of Soot Formation", Dept. of Chemical Engineering, Massachusetts Institute of Technology, Cambridge, MA, March 9, 1984.

"Soot Formation in Diffusion Flames", Workshop on Flame Radiation and Soot, Fire Research Center, National Bureau of Standards, Gaithersburg, MD, May 10, 1984.

2. By F.A. Williams

March 2, 1983, Invited Lecture, "Theoretical Aspects of Turbulent Combustion", University of California, Irvine, CA.

March 14, 1983, Invited Lecture, "Turbulent Reacting Flows", University of Texas, Austin, TX.

May 2, 1983, Invited Lecture, "Theory of Premixed Turbulent Flame Propagation", New York University, New York, NY.

August 12, 1983, Invited Lecture, "Regimes of Premixed Turbulent Flame Propagation", Air Force Office of Scientific Research, Washington, D.C.

November 12, 1983, Invited Lecture, "Structure and Propagation of Premixed Turbulent Flames", Tokyo University, Tokyo, Japan.

B. Consultative and Advisory

Liason with other A.F.O.S.R. contractors has been maintained. Further reports and information on soot formation have been supplied to the Air Force Engineering and Services Center, Tyndall AFB and AFAPL.

At the request of the Air Force System Command, Prof. Glassman served on a committee to evaluate the combustion research at the Aeropropulsion Laboratory at Wright-Patterson AFB.

Request for information with respect to soot formation have been received from Air Force Equipment suppliers such as McDonnell-Douglass, Pratt and Whitney, General Motors, Parker-Hannifin, etc. Request for publications produced under the program have been received from the following:

T.A. Litzinger, K. Brezinsky and I. Glassman, "Some Further Results on the Toluene Oxidation Mechanism", Eastern Section: The Combustion Institute, Paper No. 69, December 1982.

Beaudet, Service Recherche, Belgium
Vanpee, Univ. of Massachusetts
Sloane, GM
Tsang, NBS

C. Venkat, K. Brezinsky and I. Glassman, "High Temperature Oxidation of Aromatic Hydrocarbons", 19th Symposium Int'l on Combustion, The Combustion Institute, Pittsburgh, PA, p. 143 (1982).

Bittner, MIT
Weston, BNL
Brenner, BNL
Asaba, Univ. of Tokyo
Vandooenen, Univ. of Louvain, Belgium
Nemeth, Inst. Sci. Org. & Info., Hungary
Ludington, Purdue
Tsang, NBS
Frenklach, LSU
Semerjian, NBS
Ogunsola, Univ. of Pennsylvania
Mongia, Garrett Turbine Engine Co.
Zinn, Georgia Tech.
Walsh, MIT
Chou, Exxon
Nelson, NRL
Jachimowski, NASA Langley
Jackson, Purdue
Cernansky, Drexel Univ.
Schmieder, Sandia
Matula, LSU
Bhaskaran, IIT, India
Maly, Univ. Stuttgart
Colket, UTRC
McLean, Sandia
Calcote, AeroChem

I. Glassman, F.A. Williams and P. Antaki, "A Physical and Chemical Interpretation of Boron Particle Combustion", 19th JANNAF Combustion Meeting, Greenbelt, MD, October (1982).

Baughcum, Los Alamos National Laboratory

F. Takahashi and I. Glassman, "Sooting Correlations for Premixed Flames", Eastern States Section, The Combustion Institute Meeting, Paper No. 56, December 1982.

Beaudet, Service Recherche, Belgium
Vanpee, Univ. Massachusetts
Sloane, GM

Weissman, Univ. Southern California
Lakin, Celanese
Koser, Inst. Chem. Tech., W. Germany
Sheetz, Univ. of Dayton Research Inst.

P.A. Libby and F.A. Williams, "Strained Premixed Laminar Flames Under Nonadiabatic Conditions", Comb. Sci. and Tech. 31, 1 (1983).

I. Glassman, "Soot Reduction in Power Plants", Princeton University, Department of Mechanical and Aerospace Engineering Report No. 1632, November 1983 (AFOSR TR).

Martel, WPAFB
Soloman, Advanced Technology
Fendell, TRW

F. Takahashi and I. Glassman, "Interpretation of Sooting Correlations under Premixed Conditions", Eastern States Section, The Combustion Institute Meeting, Paper No. 12, November 1983.

Serageldin, Michigan Tech. University
Beaudet, Service Recherche, Belgium

G. Sidebotham and I. Glassman, "Sooting Behavior of Cyclic Hydrocarbons in Laminar Diffusion Flames", Eastern States Section, The Combustion Institute Meeting, Paper No. 61, November 1983.

Beaudet, Service Recherche, Belgium

T.A. Litzinger, K. Brezinsky and I. Glassman, "Some Further Results on the Oxidation of Ethylbenzene", Eastern States Section: The Combustion Institute Meeting, Paper No. 3, November 1983.

Beaudet, Service Recherche, Belgium

P.A. Libby, A. Linan and F.A. Williams, "Strained Premixed Laminar Flames with Nonunity Lewis Number", Comb. Sci. and Tech. 34, 257 (1983).

F. Takahashi and I. Glassman, "Sooting Correlations for Premixed Flames", Comb. Sci. and Tech., 33, 1 (1984).

Kraemer, Avco Lycoming

K. Brezinsky, T.A. Litzinger and I. Glassman, "The High Temperature Oxidation of the Methyl Side Chain of Toluene", Int'l J. Chem. Kinetics (in press).

Westbrook, LLL
Ravishankara, Georgia Tech.

A. Gomez, G. Sidebotham and I. Glassman, "Sooting Behavior in Temperature Controlled Laminar Diffusion Flames", Combustion and Flame (in press).

Beaudet, Service Recherche, Belgium

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Mongia, Garrett Gas Turbine Co.
Olson, AeroChem
Bittner, MIT
Sloane, GM
Coniglio, Sandia
Tsang, NBS
Scheetz, Dayton Research Inst.
Koser, Inst. Chem. Tech., W. Germany
Lakin, Celanese
Weissman, Univ. of Southern California

K. Brezinsky, E.J. Burke and I. Glassman, "The High Temperature Oxidation of Butadiene", accepted for publication in 20th Symp. Int'l on Combustion.

Sheinson, NRL
Frenklach, LSU
Kern, Univ. of New Orleans
Westbrook, LLL
Churchill, Univ. of Pennsylvania
Ravishankara, Georgia Tech.

I. Glassman, F.A. Williams and P. Antaki, "A Physical and Chemical Interpretation of Boron Particle Combustion", accepted for publication in 20th Symp. Int'l on Combustion.

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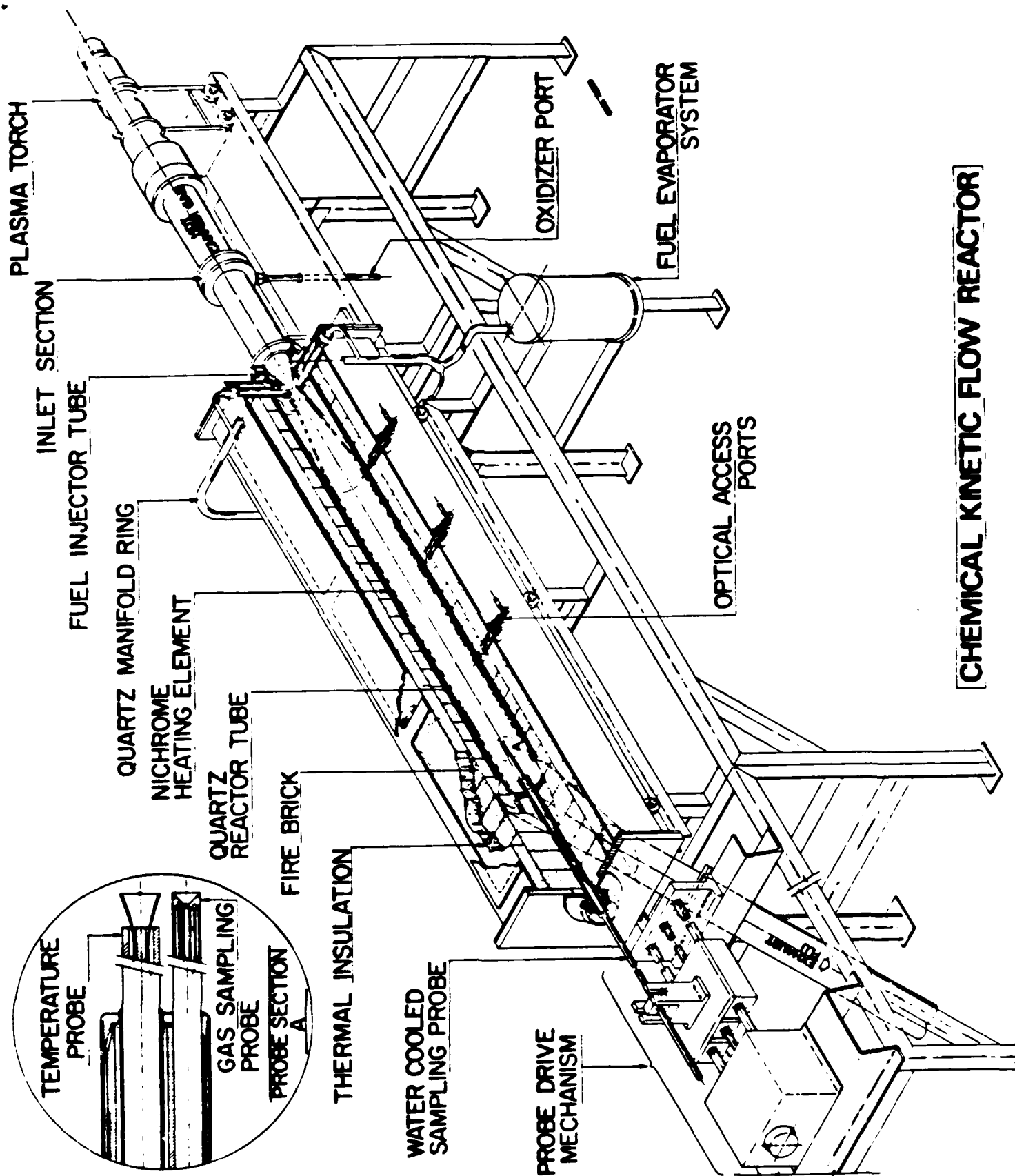


Figure 1

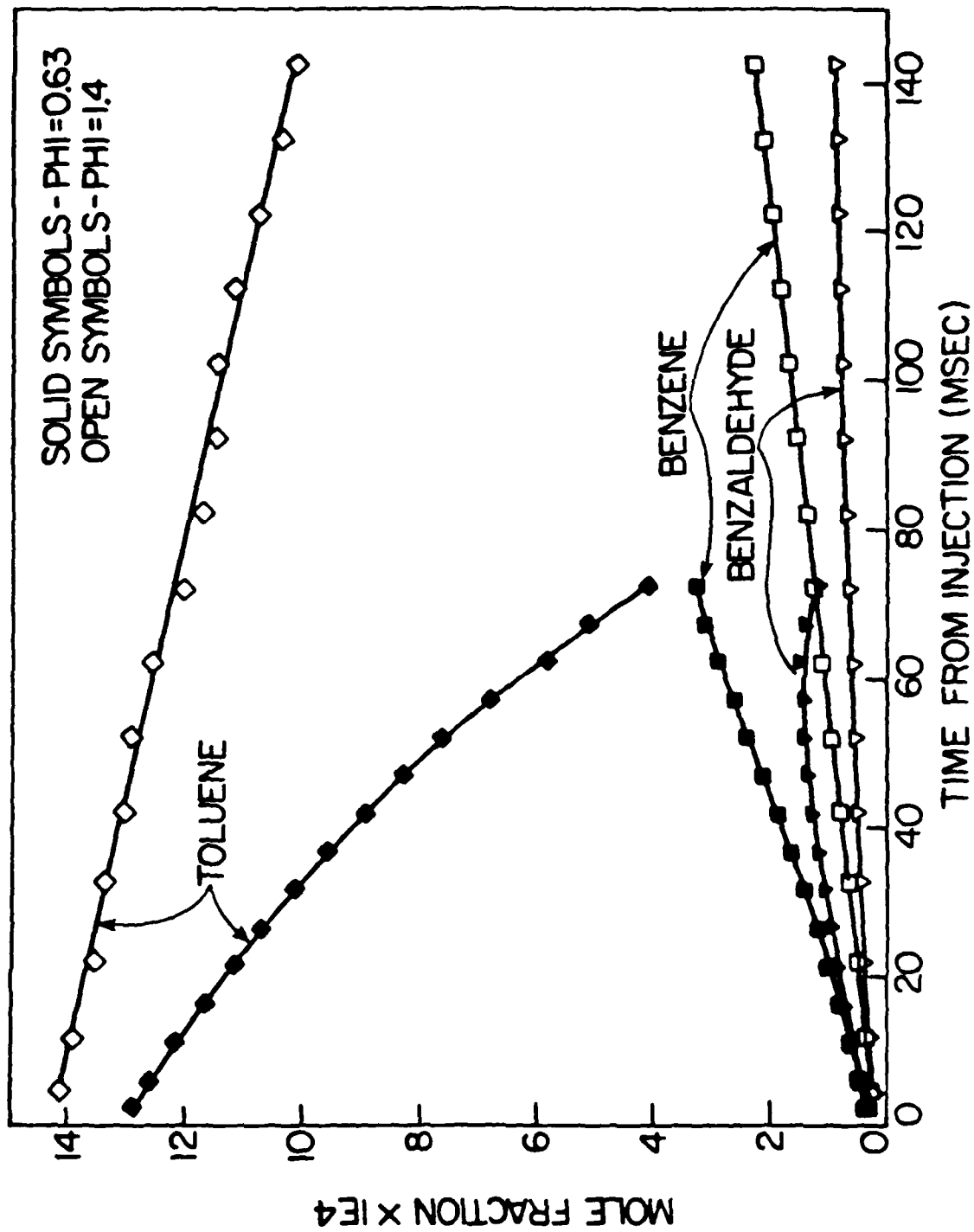


Figure 2

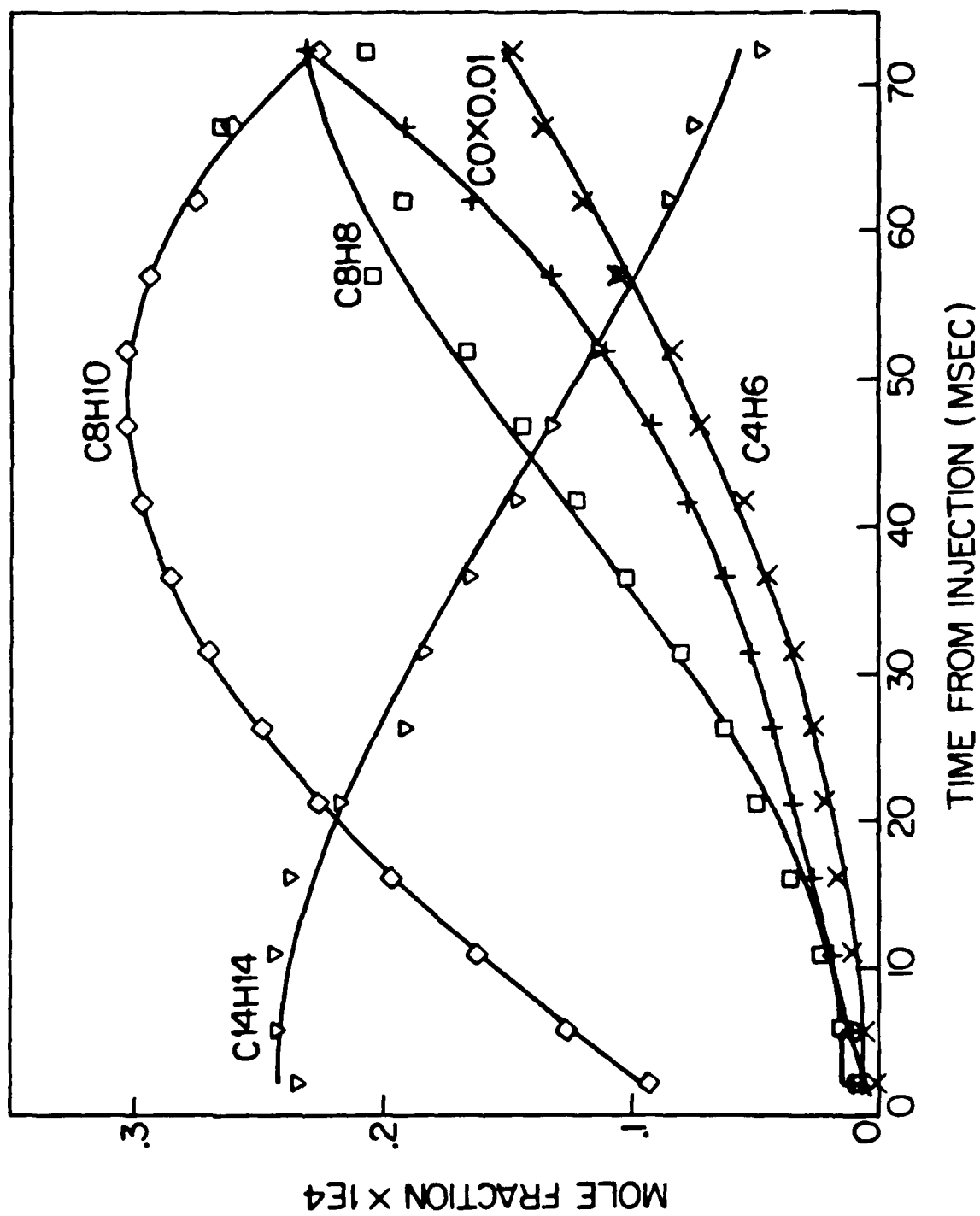


Figure 3

ETHYLBENZENE OXIDATION - $PHI=0.64$

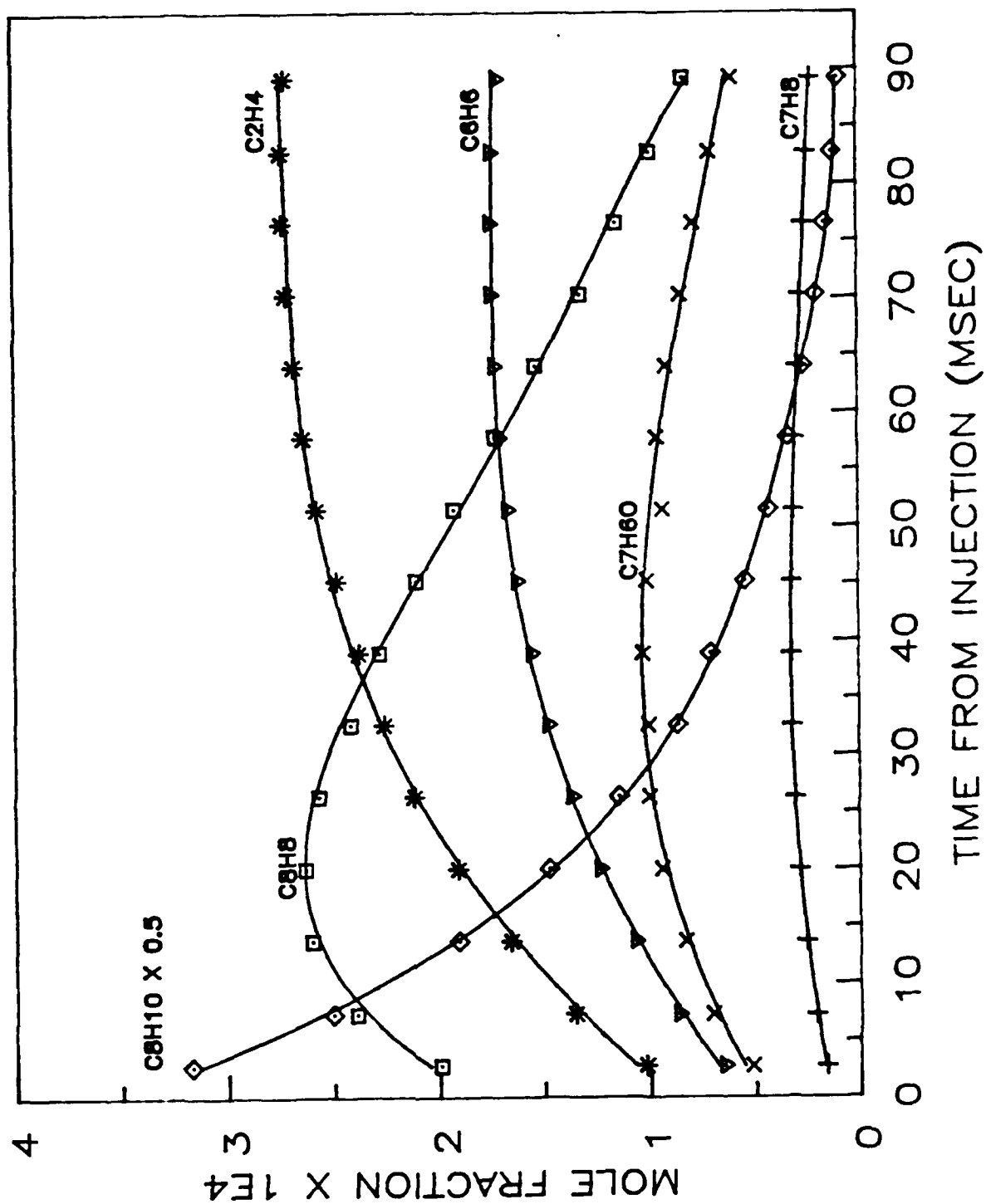


Figure 4

ETHYLBENZENE OXIDATION - $\text{PHI}=0.64$

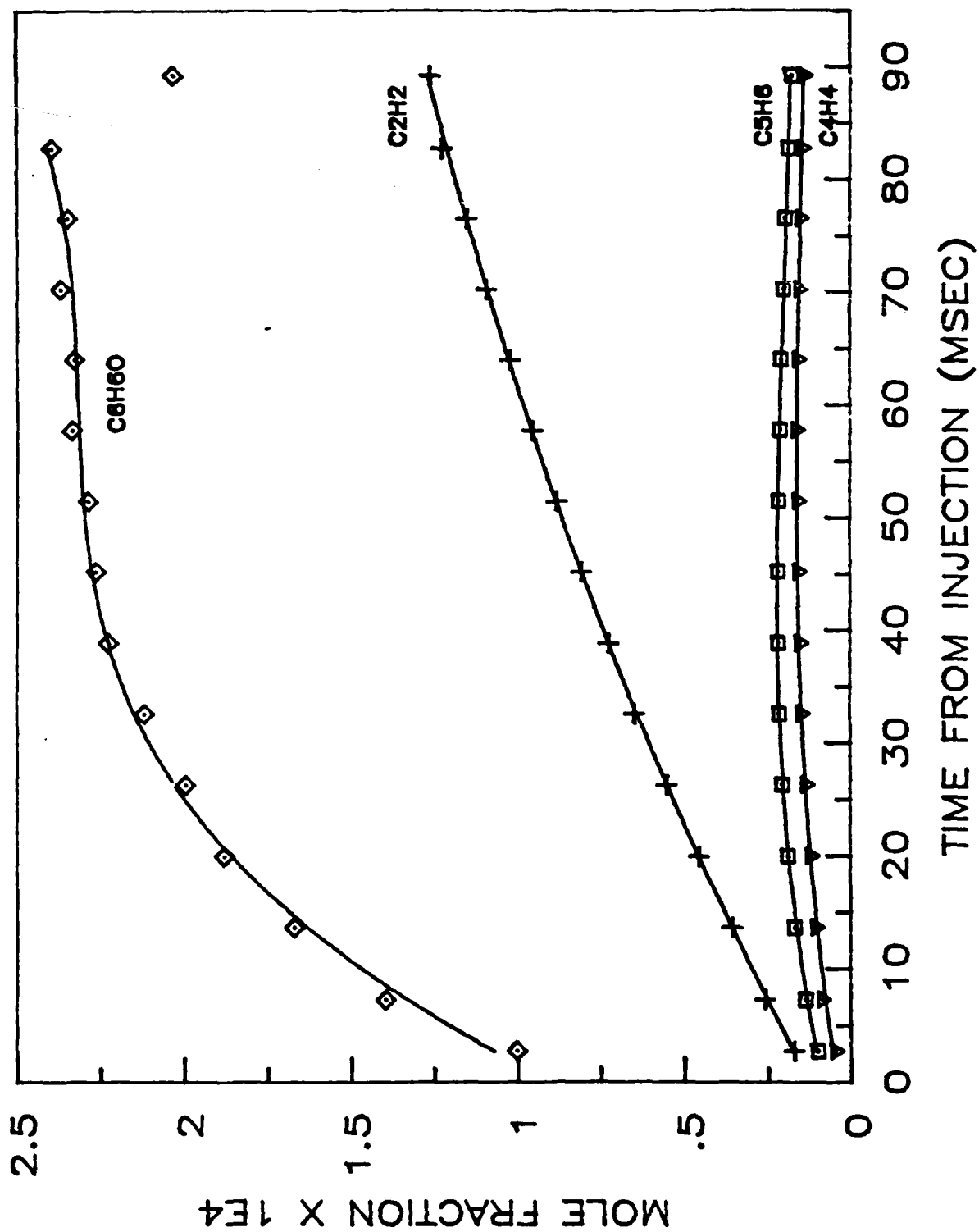


Figure 5

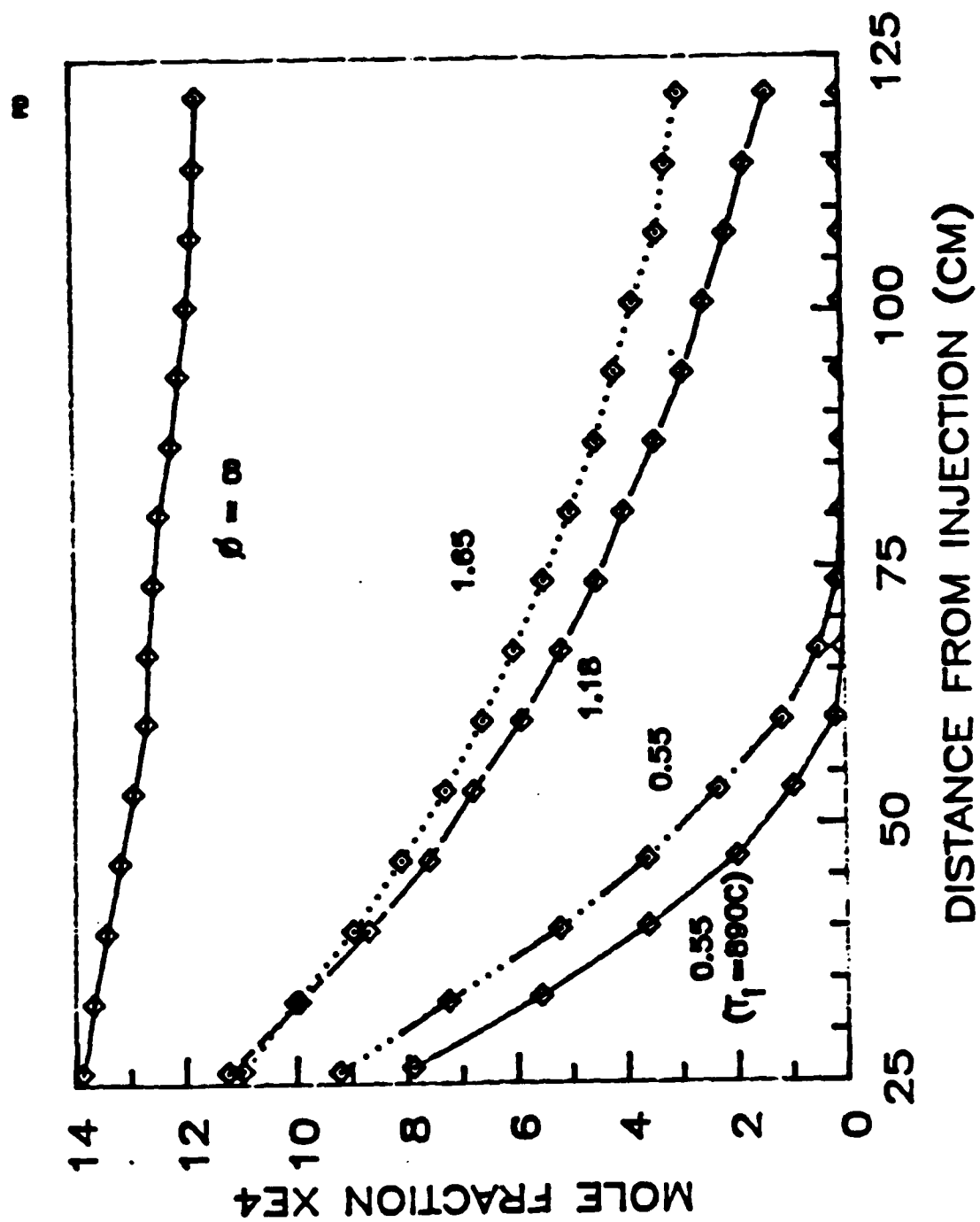


Figure 6

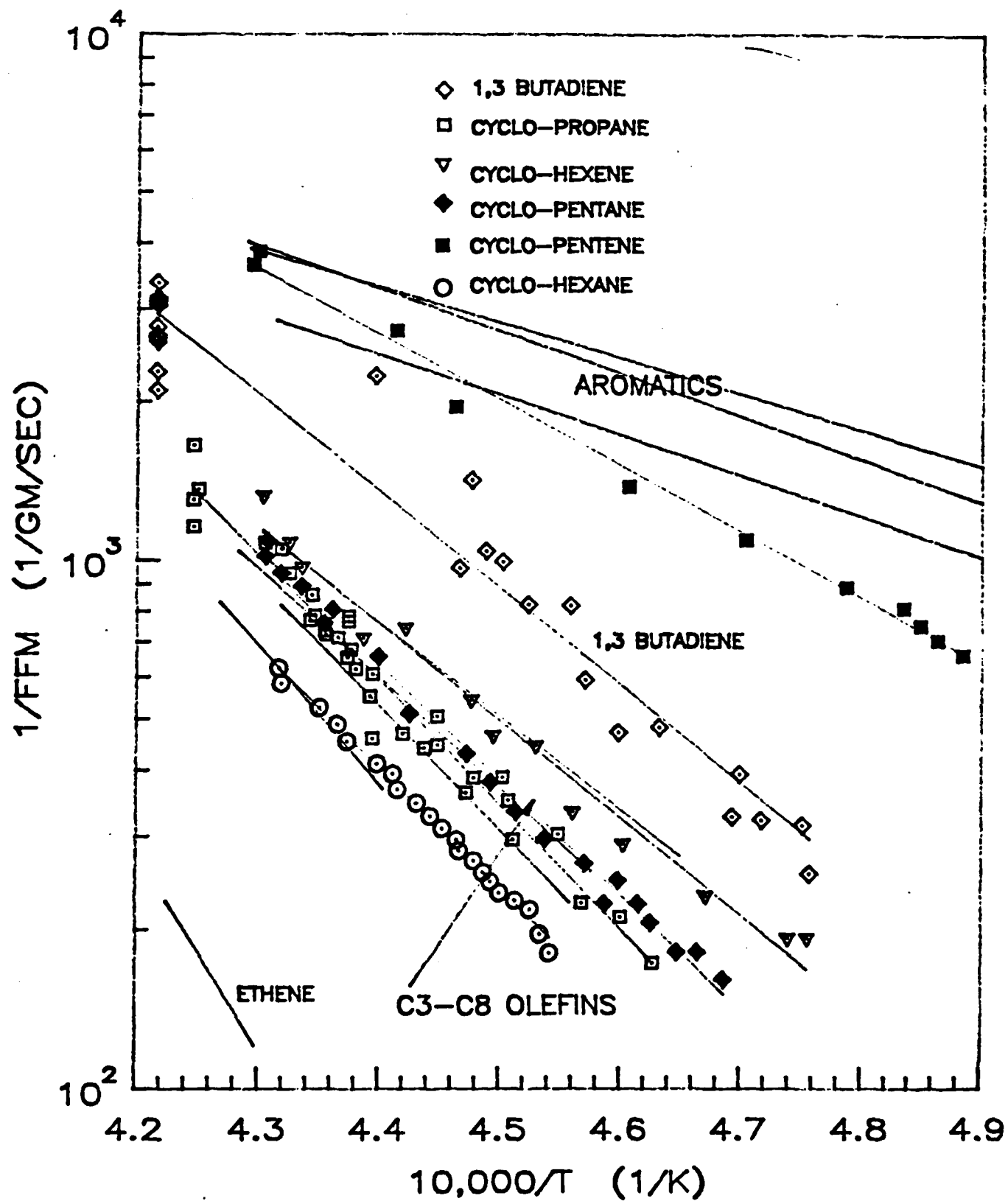


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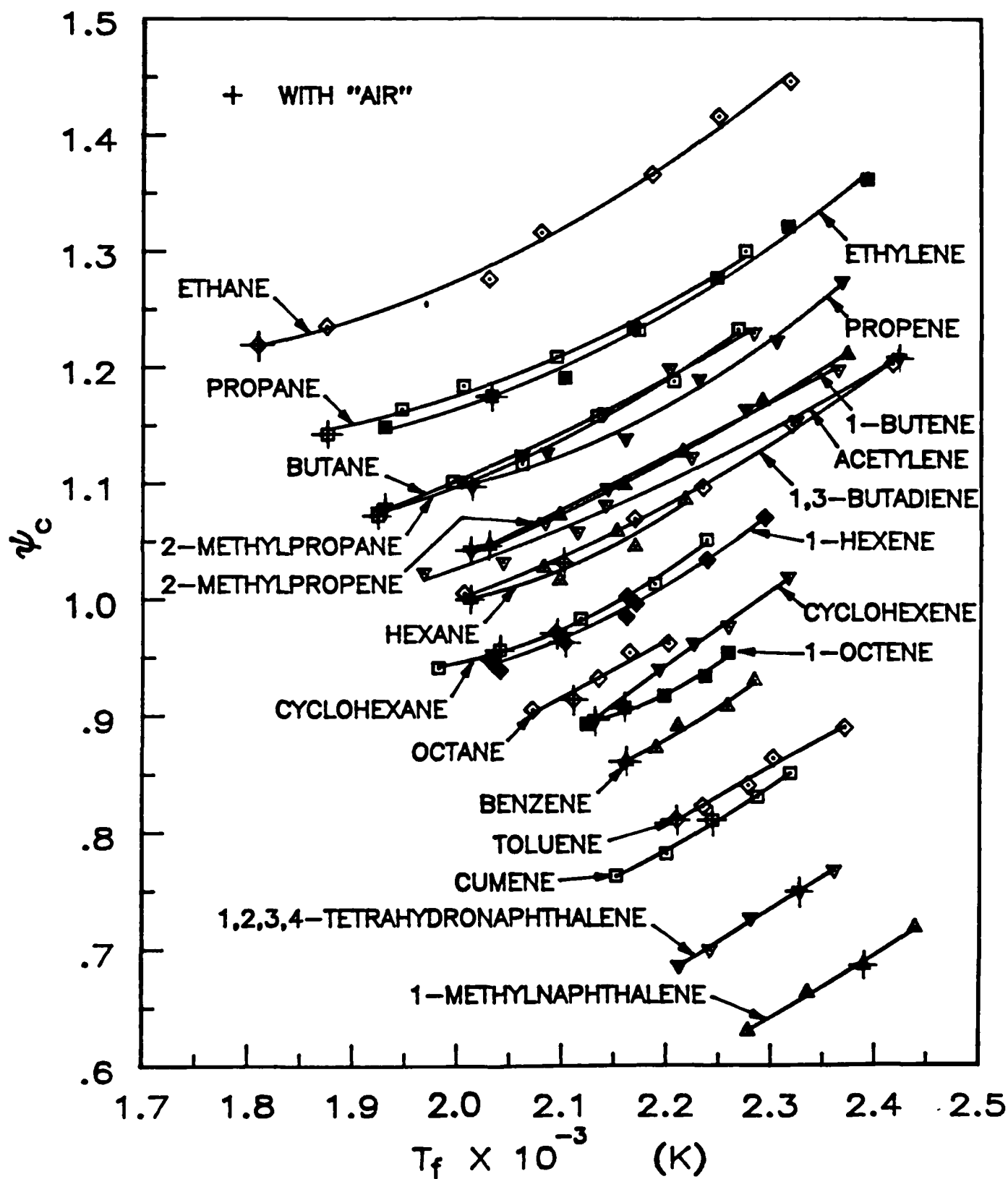


Figure 8

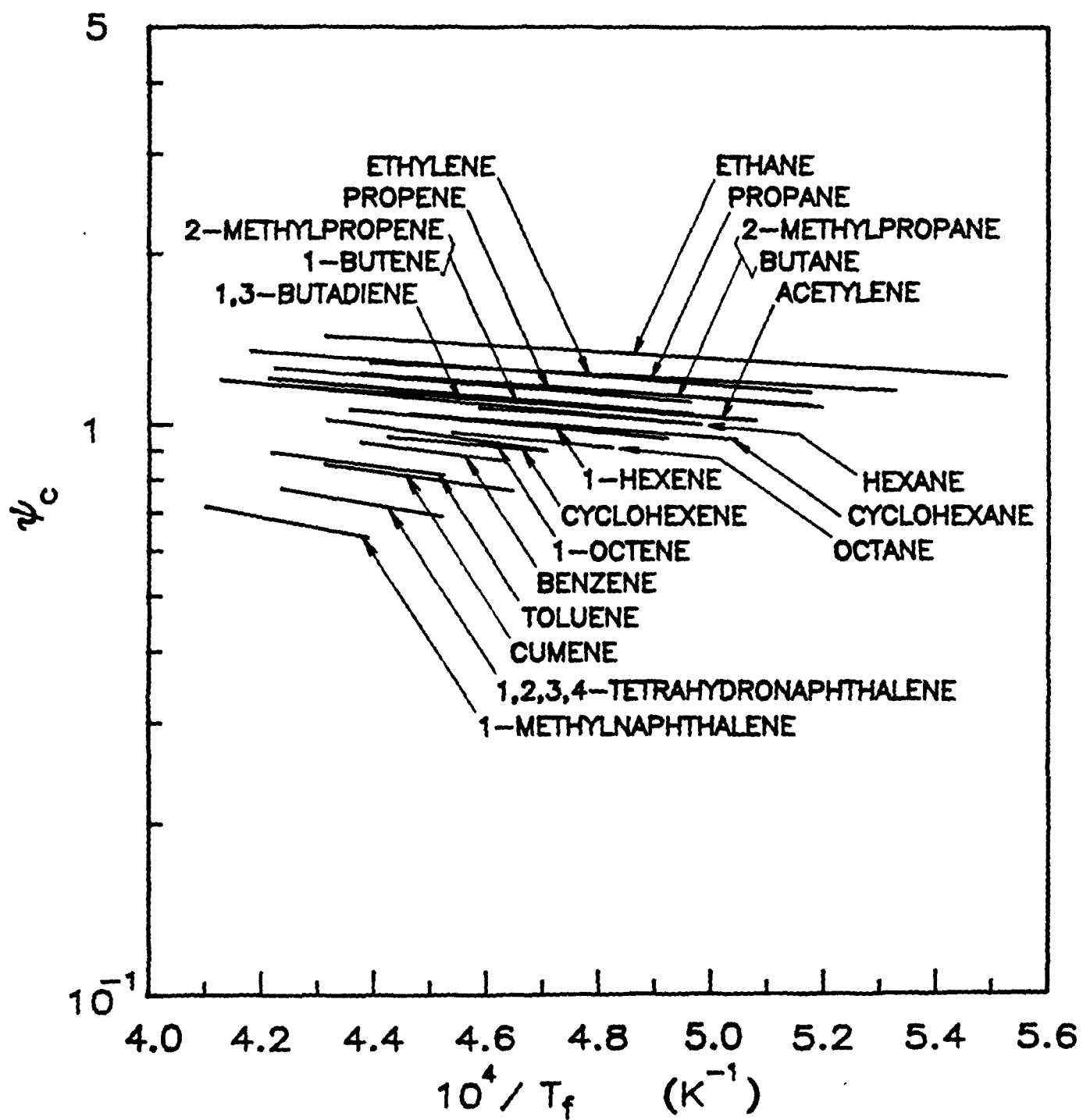


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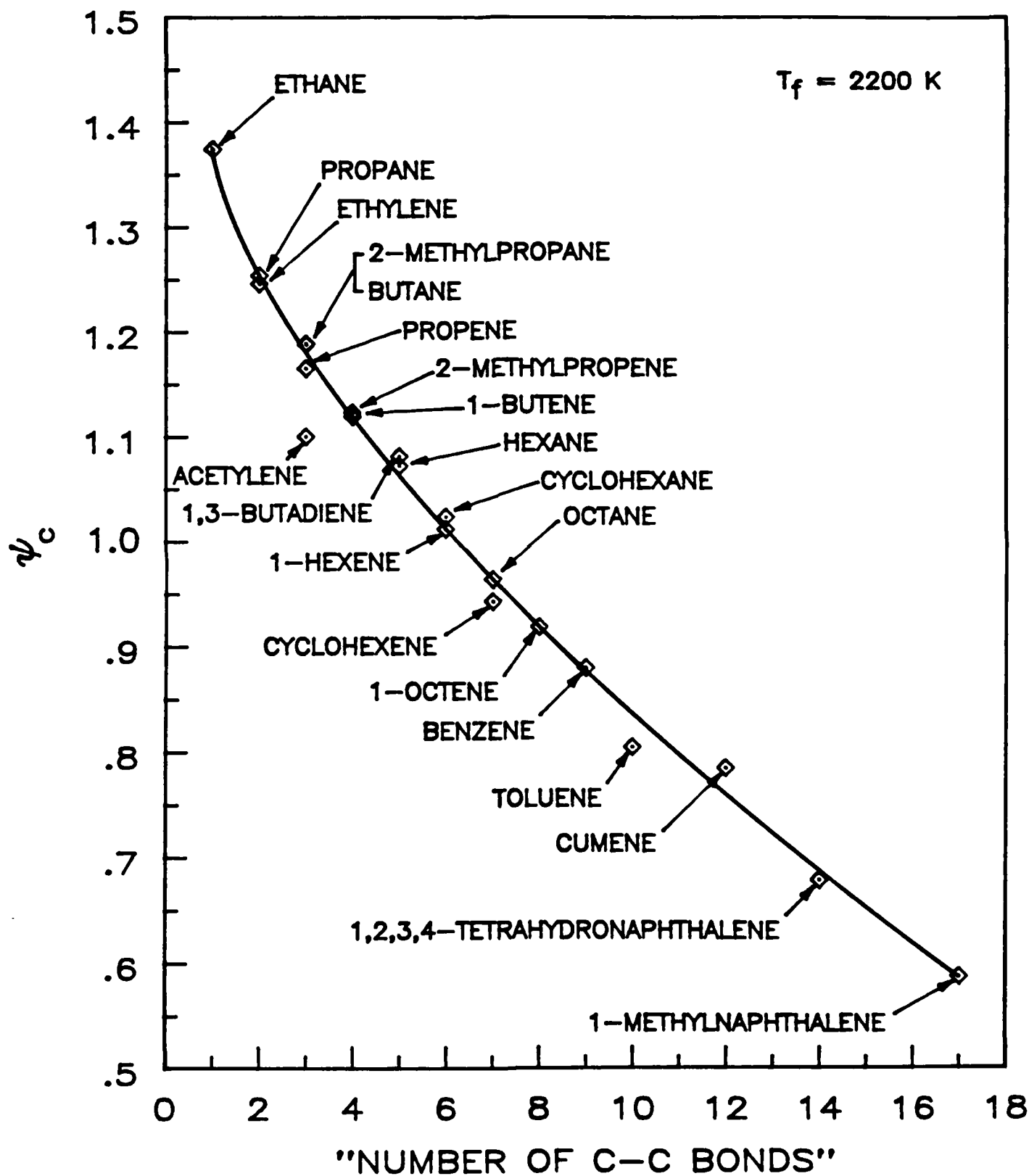


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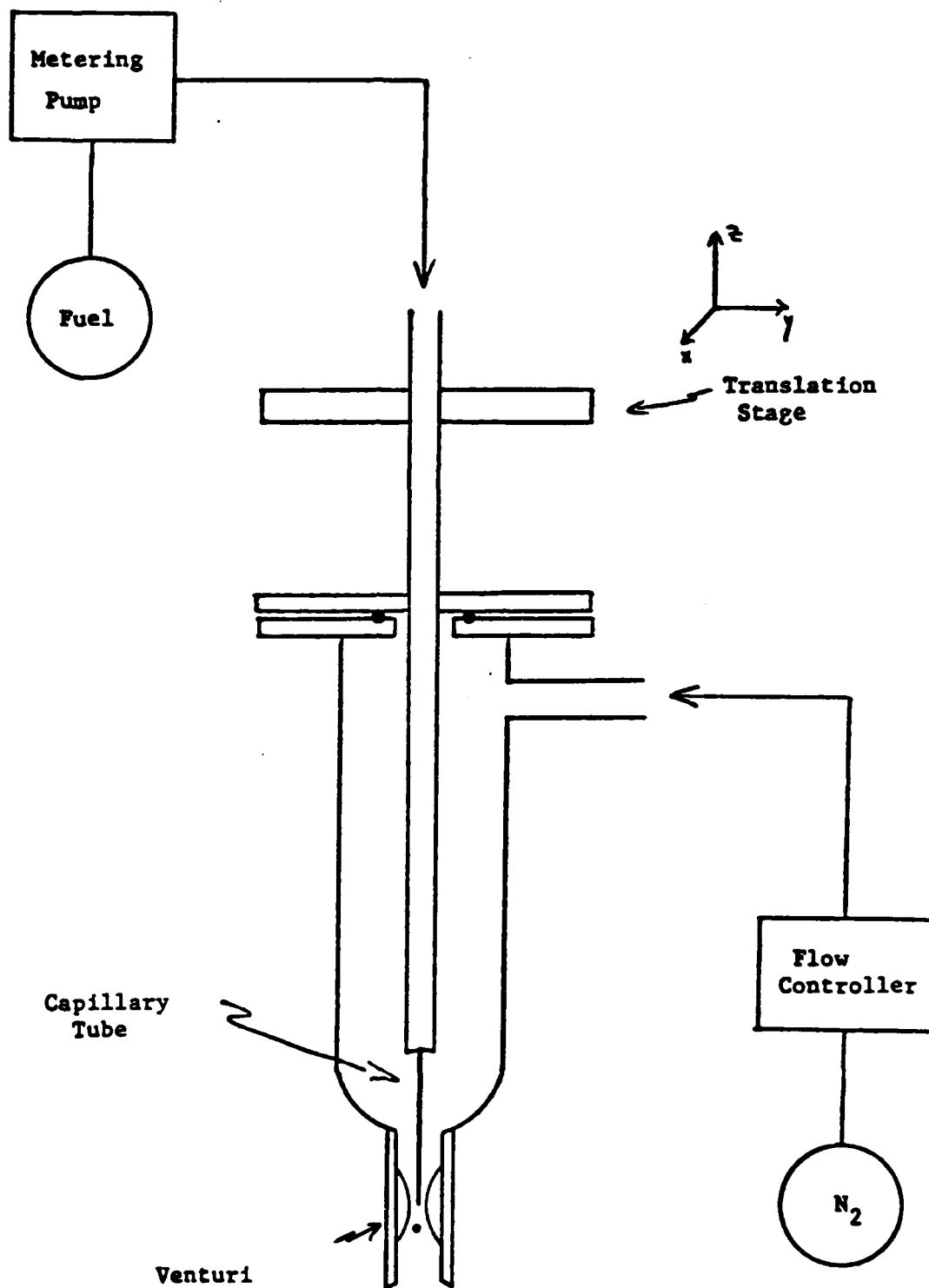


Figure 11
SCHEMATIC OF DROPLET DEVICE

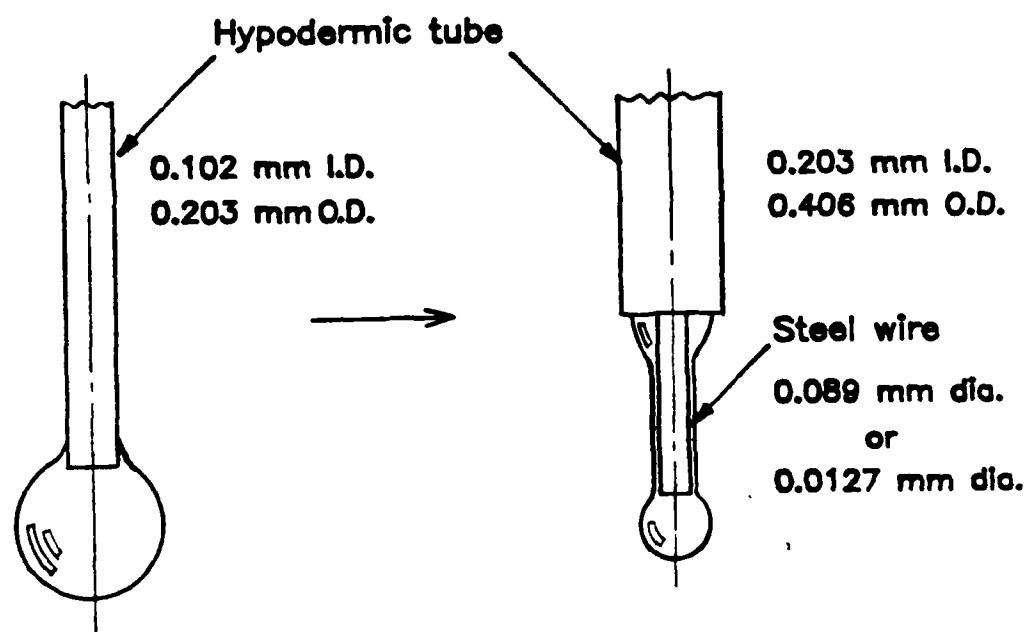


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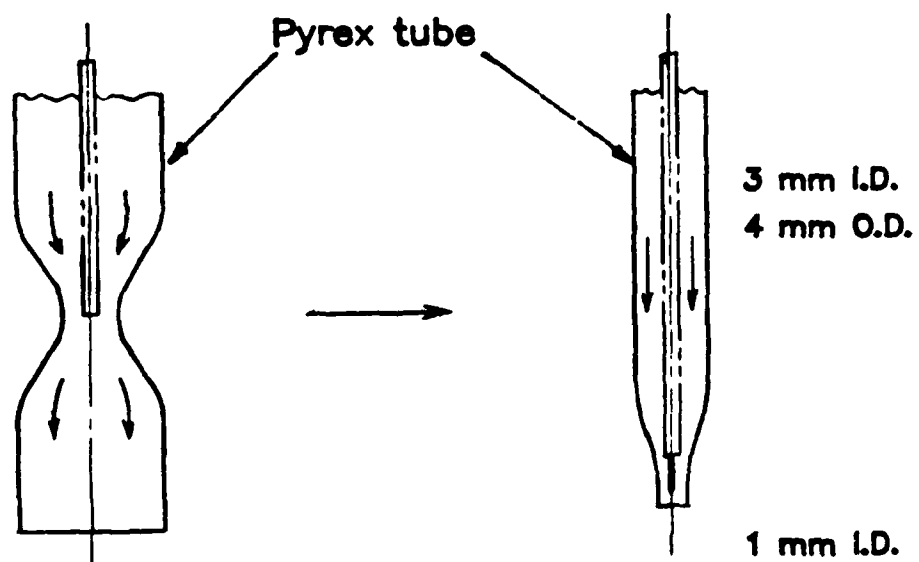


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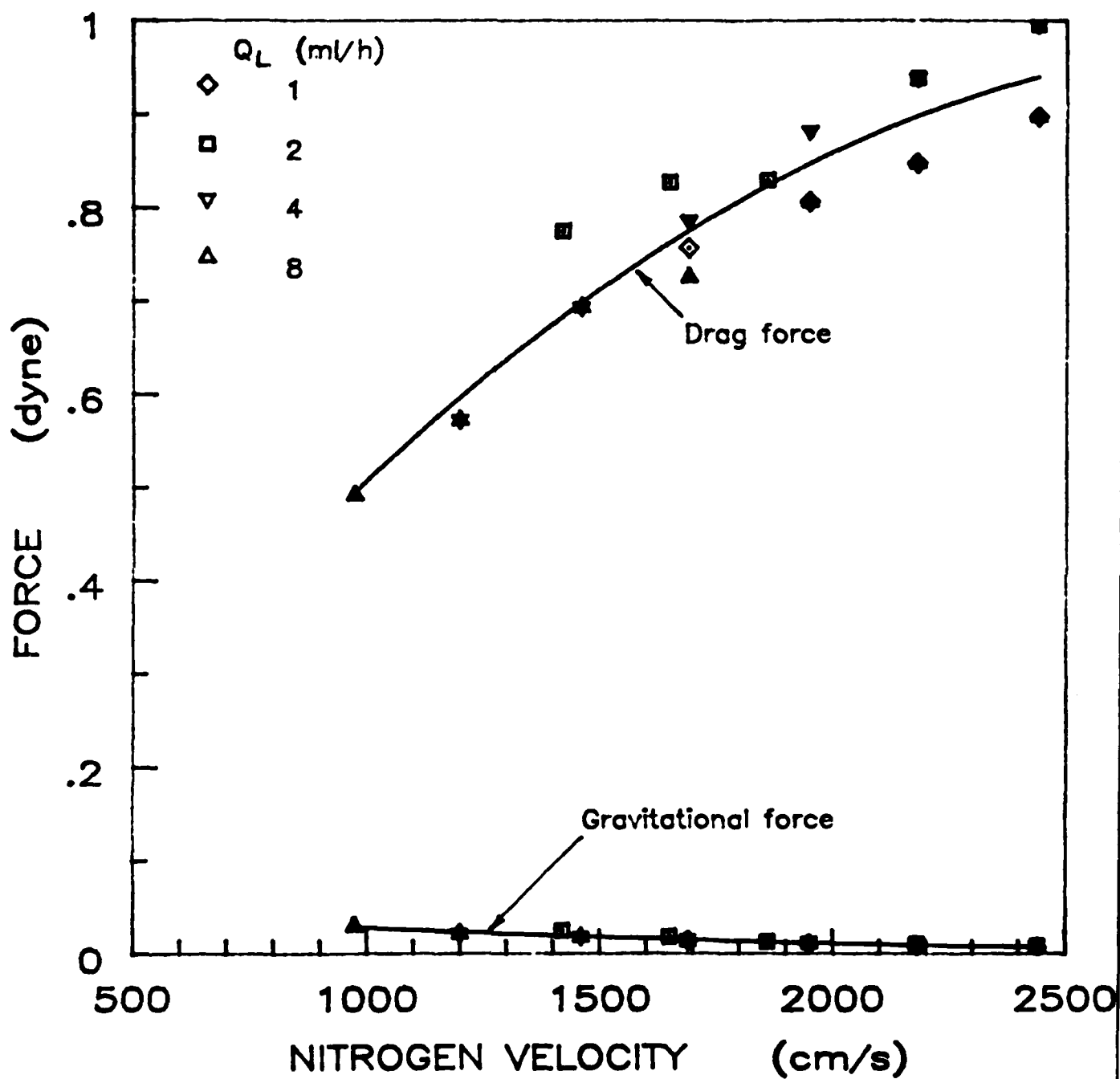


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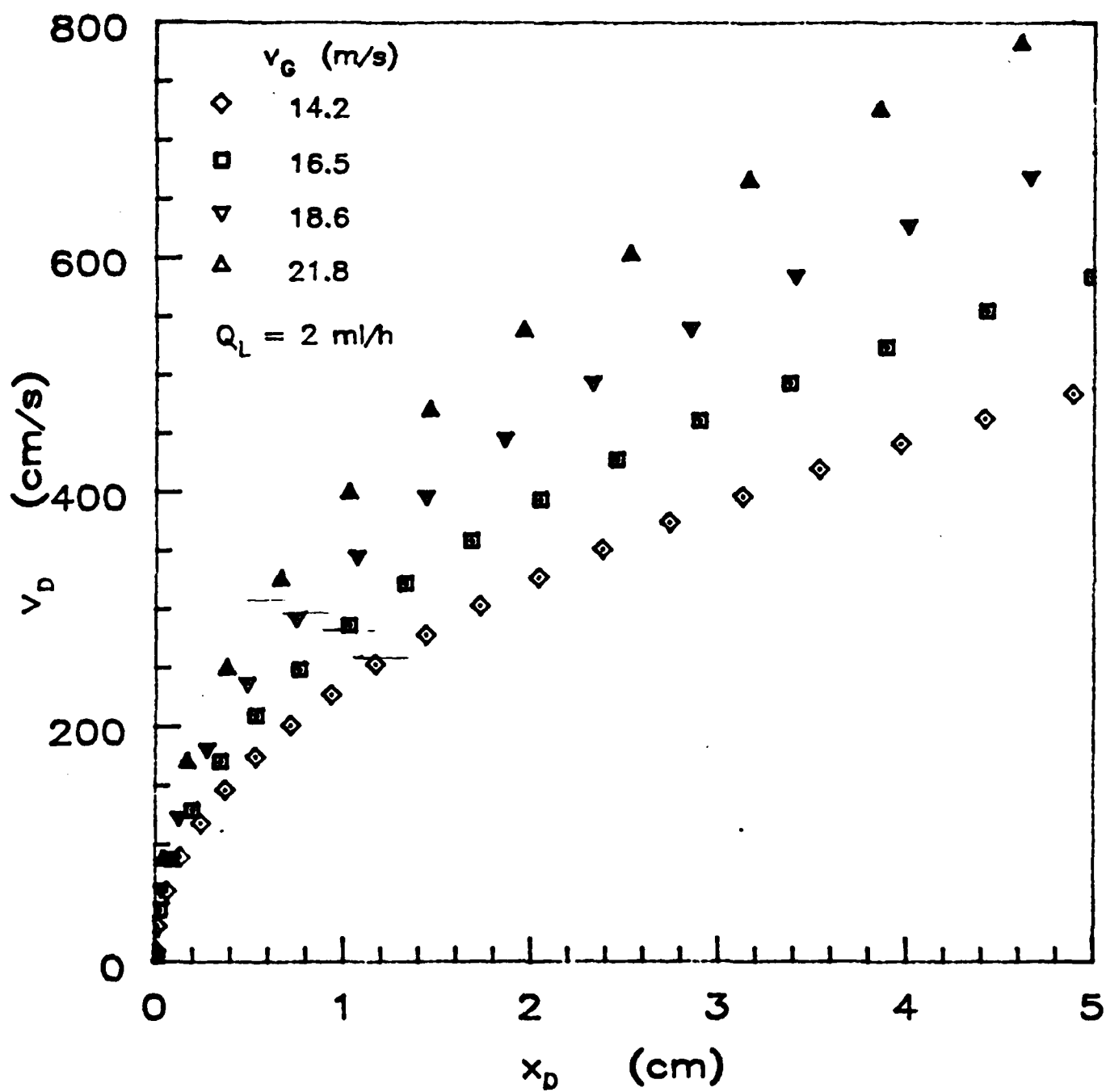
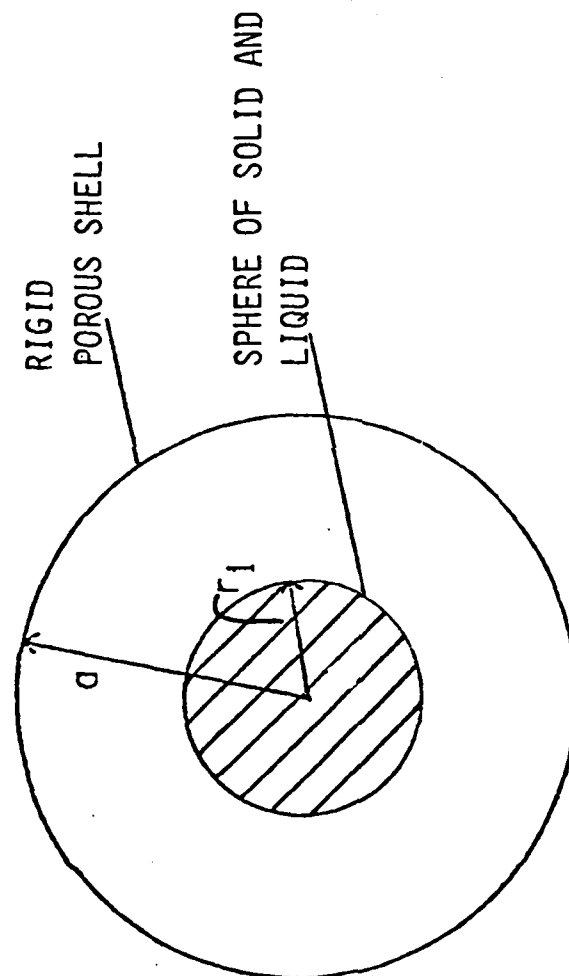


Figure 15



a = constant outer radius of porous shell
 r_1 = decreasing radius of solid and liquid sphere
 POROUS SHELL MODEL AFTER SIGNIFICANT EVAPORATION
 OF LIQUID

Figure 16

END

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